P 12 Sparse optimal control in time-dependent density functional theory (G. Friesecke, K. Kunisch) → NS, IS

This project is motivated by application problems of current interest in quantum control, which range from steering chemical reactions [1] over creating excited or ionized states [7] to faithfully storing and manipulating bits of quantum information [12]. The goal of the project is to develop a mathematical theory and numerical implementation of frequency-sparse optimal control for time-dependent density functional theory (TDDFT). This will advance the two goals of simple controls, not reached in traditional simulations, and applicability to large systems, which motivates our focus on TDDFT which replaces the linear but high-dimensional Schrödinger equation by a non-linear but low-dimensional system which poses interesting mathematical challenges.

State of the art. While control fields for quantum systems designed by standard optimal control methods have found important applications, they typically exhibit a complicated time-frequency-structure and are not easy to interpret and implement in experiments. A novel approach which results in controls with a much simpler and sparse time-frequency-structure was recently developed within the IGDK by the project proposers in collaboration with their joint PhD-student F. Henneke [8]. This approach is built upon the basic idea of minimizing sparsity-enhancing, L^1- or measure-norm cost functionals as introduced in the context of elliptic and parabolic problems in [13, 10]. More specifically, it combines the recent idea of optimization with function-valued measures to achieve directional sparsity in parabolic control [11] with the idea that in quantum control, unlike in parabolic control, the target for effectiveness and sparsity should not be the field amplitude but its frequency structure.

Hitherto, the mathematical theory of frequency-sparse optimal quantum control has only been developed for "exact" (linear) quantum dynamics; but due to the steep scaling of computational cost for computing quantum wavefunctions (functions on $\mathbb{R}^{3N}$ for N particles) the implementation has been limited to small systems. On the other hand, time-dependent density functional theory (TDDFT) [3] offers a reasonably accurate and widely used model for simulating quantum dynamics of large systems, by replacing the linear evolution of the high-dimensional wavefunction by a nonlinear system for N single-particle wavefunctions on $\mathbb{R}^3$. This fact is appreciated in the physics literature and since about 2010 interest is emerging in combining optimal control with TDDFT (see e.g. [4, 7]). But at present a mathematical theory for the TDDFT control problems with classical costs considered in the physics literature is not available, and sparsity-promoting costs as proposed here appear to have not been considered at all.

Thesis project to be supervised by Gero Friesecke. We seek to develop a mathematical framework and numerical implementation for generating frequency-sparse controls in time-dependent density functional theory (TDDFT). Mathematically, this corresponds to analyzing and simulating a coupled system of nonlinear partial differential equations of form

$$i\partial_t\varphi_i = \Big(-\frac{1}{2}\Delta + v_{ext} + \frac{1}{|\cdot|} * \rho(\cdot,t) + v_{xc}[\rho] + (Bu)(t)D\Big)\varphi_i \quad (i = 1,..,N),$$

$$\rho(x,t) \equiv \sum_{i=1}^{N} |\varphi_i(x,t)|^2$$

for N functions $\varphi_i : \mathbb{R}^3 \times [0,T] \to \mathbb{C}$ and determining the control field $u : \Omega \times [0,T] \to \mathbb{C}$ by minimization of a tracking term plus a cost term,

$$-\sum_{i=1}^{N} \langle \varphi_i(T), P\varphi_i(T) \rangle \quad + \quad \alpha \|u\|.$$

Here P is the projector onto a target subspace, $(Bu)(t)$ is a laser field amplitude, the control $u(\omega, t)$ is a suitable time-frequency decomposition of the laser field with reconstruction operator B [8], and $\|\cdot\|$ is a function-valued measure norm which promotes sparsity in frequency and smoothness in time. The φ_i are the one-body Kohn-Sham orbitals of the many-electron system, ρ is the electron density, D a dipole operator, v_{ext} the Coulomb potential exerted on the electrons by the atomic nuclei, and $v_{xc}[\rho]$ an exchange-correlation potential which models many-body effects and depends on the electron density ρ [3]. A simple example (exchange-only local density approximation, TDLDA) is $v_{xc}[\rho] = -\text{const}\,\rho^{1/3}$. Natural other examples are exact exchange, which becomes exact in a weak interaction limit, and the Kantorovich potential from optimal transportation theory associated with optimal placement of electrons, which becomes exact in a strong interaction limit [6, 5].

The first step in the project will be to adapt the standard existence theory for bilinear optimal control problems [2] to the above nonlinear system. The novel task is to deal with the nonlinear coupling terms $|\cdot|^{-1} * \rho$ and $v_{xc}[\rho]$ in the evolution system. The next goal is to derive and analyze the optimality system by combining structural features of TDDFT such as orbital orthonormality preservation and regularity propagation with recent insights on optimal control with function-valued measure norms [11]. To numerically solve the optimality system we plan to combine previous work in our group [9] on quantum dynamics in the context of optimal control, which appears flexible enough to accommodate the nonlinear coupling, with appropriate optimization routines such as L-BFGS which bypass the expensive computation and storage of Hessians. Our starting point for handling the nonlinear coupling will be the iterative methods in the physics literature for bare TDDFT evolution. These alternate between updating the Kohn-Sham orbitals via linear evolution at frozen density and updating the density from the new Kohn-Sham orbitals, and should be adaptable to including the backward evolution for the dual state. Finally we will use our implementation to generate sparse controls for model problems such as those simulated in [7], and conduct numerical experiments to investigate the important theoretical question how the choice of exchange-correlation potential affects the structure and yield of the optimal controls.

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