1. Problem Description

Set $E \leq E_{\text{fin}}(t=0)$ should be mapped to set $E \geq E_{\text{fin}}(T)$ with time evolution operator $\hat{U}(T,0;\mathbf{c}(t))$.

$\hat{U}$: operator that maps state according to equation of motion, propagated from 0 to T.

TDSE: $i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = \hat{H}(\mathbf{c}(t)) |\psi(t)\rangle$; e.g. $\hat{H} = \begin{pmatrix} 0 & V_0 e^{\mathbf{c}(t)} \\ V_0 e^{\mathbf{c}(t)} & 0 \end{pmatrix}$.

What is the control that brings the system from its initial state to its target state?

2. Discretization

control is piecewise constant: switches at points of time grid

$\hat{U}(T,0;\mathbf{c}(t)) = \prod_{i=0}^{N-1} e^{i \mathbf{c}(t_i) \hat{H}} = \hat{U}_N \ldots \hat{U}_2 \hat{U}_1 |\psi(0)\rangle$

Propagation: Runge-Kutta or Chebyshev if more precision is required.

3. Optimization Functionals

OCT: minimize a functional of the control $\mathbf{c}$ includes measure of success, possibly penalties (cost functional) for the control and/or the system evolution.

iterative procedure: start with guess $\mathbf{c}^{(0)}(t)$, find improvement

$\mathbf{c}^{(n)}(t) = \mathbf{c}^{(0)}(t) + \Delta \mathbf{c}(t)$, repeat.
Measure of success: Fidelities

\[ \rho_{\text{true}}^{(t)} \xrightarrow{} \rho_{\text{true}}^{(t+\Delta t)} \]

Fidelity is functional of states at final time \( T \)

Add intermediate-time constraints for full optimization functional:

\[ \delta \rightarrow 0 \quad \text{as} \quad t \rightarrow 0 \]

4. Motive - Method


Specify \( J(t) = 2\alpha \int_{t_0}^{t_1} \dot{E}(t) \, dt + 2\lambda \int_{t_0}^{t_1} \dot{E}(t) \, dt \)

Optimization Equation

Minimize \( J \) by choosing control \( \epsilon \), but such that dynamics of states follows eq. of motion

Idea of motive: auxiliary functional with additional arb. potential \( \Phi \), use freedom in \( \Phi \) to construct improved field
\[ S = \mathcal{S}(\mathcal{E}(\eta(t))) + \int_0^T g_o \frac{e(t)}{\mathcal{E}(\eta(t))} \, dt + \int_0^T g_b \mathcal{R}(\mathcal{E}(\eta(t))) \, dt \]

\[ L = \mathcal{S}(\mathcal{E}(\eta(t))) - \Phi(\mathcal{E}(\eta(t)), 0) - \int_0^T \mathcal{R}(\mathcal{E}(\eta(t))) \, dt \]

with\[ g = \dot{S}T[\mathcal{E} \eta(t) \mathcal{E}^T] + \Phi(\mathcal{E} \eta(t), T) \]

\[ r = - \left[ g_o \frac{e(t)}{\mathcal{E}(\eta(t))} + g_b \mathcal{E} \eta(t) \mathcal{E}^T \right] + \frac{\partial \Phi}{\partial e} + \sum_{k=1}^N \left[ \nabla \Phi \frac{\partial f_k}{\partial \eta} [\mathcal{E} \eta(t), e(t)] + \alpha \eta \nabla \Phi \frac{\partial f_k}{\partial \eta} [\mathcal{E} \eta(t), e(t)] \right] \]

\[ \text{eq. of media: } \eta_k \]

\[ \text{For any potential } \Phi : L = g \]

Expand \( \Phi \) into states:

\[ \Phi[\mathcal{E} \eta(t), e(t)] = \sum_k \left( \langle x_k(t)| \eta_k(t) \rangle + \langle \eta_k(t)| x_k(t) \rangle \right) + \text{(second order)} \]

\( \h \) maximize \( L \) with respect to the states (by choosing coefficients \( \alpha \) appropriately) \( \Rightarrow \) any change in control will then improve \( L \)

\[ \Rightarrow \text{eq. of media for } x : \]

\[ \frac{d}{dt} |x(t)\rangle = -\frac{i}{\hbar} H^+ \left( \mathcal{E}(\eta(t)) \right) |\langle x(\eta(t))| + \alpha \Delta \eta_k \delta_k \]

\[ |\langle x(\eta(t))| = -\frac{\alpha}{\Delta \eta_k} \delta_k \]

\( \text{Minimize } L \) with respect to the field (or-functional)

\[ \frac{\partial g}{\partial e} \bigg|_{\eta, \varphi_0} = \text{Im} \sum_k \left( \sum_{k=1}^N \langle x_k^{(\eta)}(t)| \frac{\partial H}{\partial e} \rangle_{\varphi_0} \right) |\eta_k^{(\eta)}(t)\rangle \]

\[ \Rightarrow \Delta e = \frac{S(t)}{\lambda_o} \text{Im} \sum_k \left( \sum_{k=1}^N \langle x_k^{(\eta)}(t)| \frac{\partial H}{\partial e} \rangle |\eta_k^{(\eta)}(t)\rangle \right) + \text{higher order} \]
Comments:

- Expanding up to second order in states \( \hat{\psi} \) may be required for first order: \( \frac{d}{dt}, g, \mu \) etc. of motion linear in states, control.

- System Hamiltonian enters as \( \frac{d}{dx} \), usually \( \frac{d}{dx} = \mu \), but other equations of motion might be used.

- Find-time functional / fidelity enters through boundary conditions for \( \psi \):

\[
F_{\text{re}} = \frac{1}{N} \text{Re} \sum_{k=1}^{N} \left( \psi_k^{+g|} \Psi_k(+) \right) = \frac{1}{N} \sum_{k=1}^{N} \frac{1}{2} \left( \left< \psi_k^{+g|} \Psi_k(+) \right> + \left< \Psi_k(+) \psi_k^{+g|} \right> \right)
\]

\[
\Rightarrow \left| \chi_k(+) \right> = \frac{1}{N} \left( \frac{1}{2} \left( \left< \psi_k^{+g|} \Psi_k(+) \right> + \left< \Psi_k(+) \psi_k^{+g|} \right> \right) \right)\left| \psi_k^{+g} \right>
\]

\[
F_{\text{im}} \Rightarrow \left| \chi_k(+) \right> = \frac{1}{N^2} \left( \sum_{k=1}^{N} \left< \psi_k^{+g|} \Psi_k(+) \right> \right) \left| \psi_k^{+g} \right>
\]

\Rightarrow \text{Slides}

\Rightarrow \text{example of application:} \ [\text{Goertz et al. J. P. B. At. Mol. Opt. Phys. 44, 154011}] ; \ [\text{www.physik.uni-kassel.de/en/koch.html}]

5. Gradient Ascent

Alternative algorithm to Hrotor \ [\text{Khaneja et al. J. Magn. Res. 172, 296 (2005)}]

\[
\text{Change pulse in direction of gradient:} \quad \varepsilon^{(a)}(t) = \varepsilon^{(0)}(t) - \alpha \frac{\partial \delta T}{\partial \varepsilon(t)}
\]
Second order (\( \approx \) Newton) is necessary to reach convergence.

LBFGSB is an algorithm that approximates Hessian from first derivatives (black-box algorithm).

**Calculation of gradient** \(- \frac{\partial J_t}{\partial E(k)}\)

Assume \( J_t = \langle \psi^T_s | \hat{U}(T, 0) | \psi^m \rangle \); \( U(T, 0) = \frac{1}{i} \hat{U}_i \)

\[
\frac{\partial \hat{U}_i}{\partial E} = \sum_{j=0}^{i} \left( \frac{\partial \hat{U}_i}{\partial E(j)} \right) \frac{1}{j!} \hat{U}_j \quad U_i = e^{-i \hat{H}(E_i) t} \Rightarrow \text{series expansion}
\]

\[
\frac{\partial \hat{U}_i}{\partial E} = \sum_{k=0}^{N} \frac{(-igt)^k}{k!} \sum_{n=0}^{\infty} \hat{H}^k \left( \frac{\partial H(E_i)}{\partial E} \right) \hat{H}^{n-k-1} \quad \text{truncate after sufficient } N
\]

**Note:**

- Kramer's sequential (\( E^{(n)} \) acts update); gradient method is not
- Convergence is not strictly guaranteed
- Different derivatives need to be calculated: \( \frac{\partial J_t}{\partial E} \) vs. \( \frac{\partial J_t}{\partial H} \).