A Time-dependent method for electronic conduction and open systems

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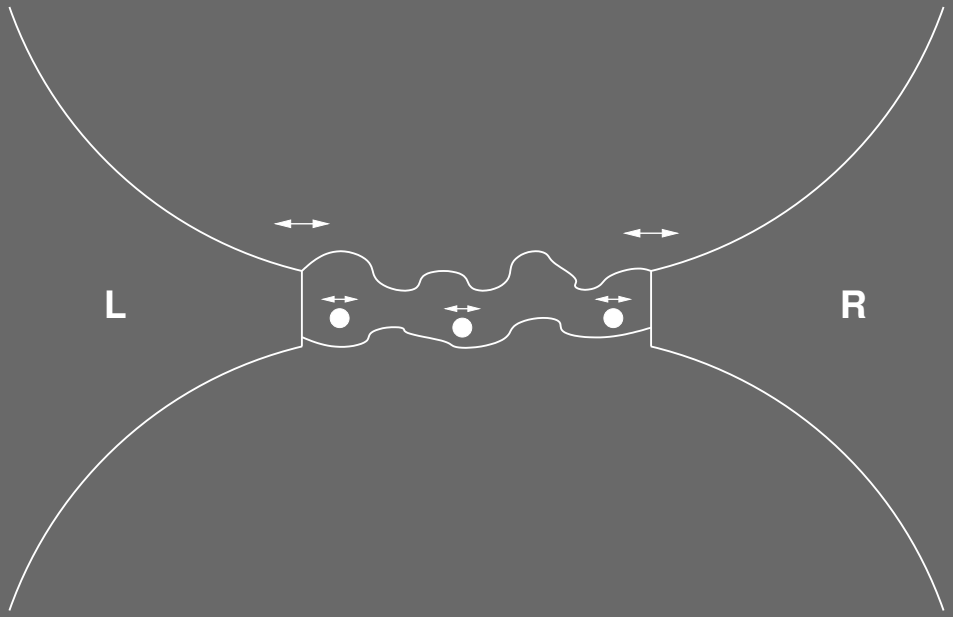
Introduction

- Landauer introduced two key ideas to the physics of meso- and microscopic conduction:
  - The field in the device arises from the build up of carriers at localised scatterers
  - Conductance arises from (can be viewed as) transmission
- We consider (as below) left electrode, device, right electrode



Introduction(2)

- We can derive the formula for conductance, G, from both the Kubo and Keldysh formalisms:

$$G = \frac{2e^2}{h}\mathrm{Tr}\left(tt^\dagger\right) \tag{1}$$

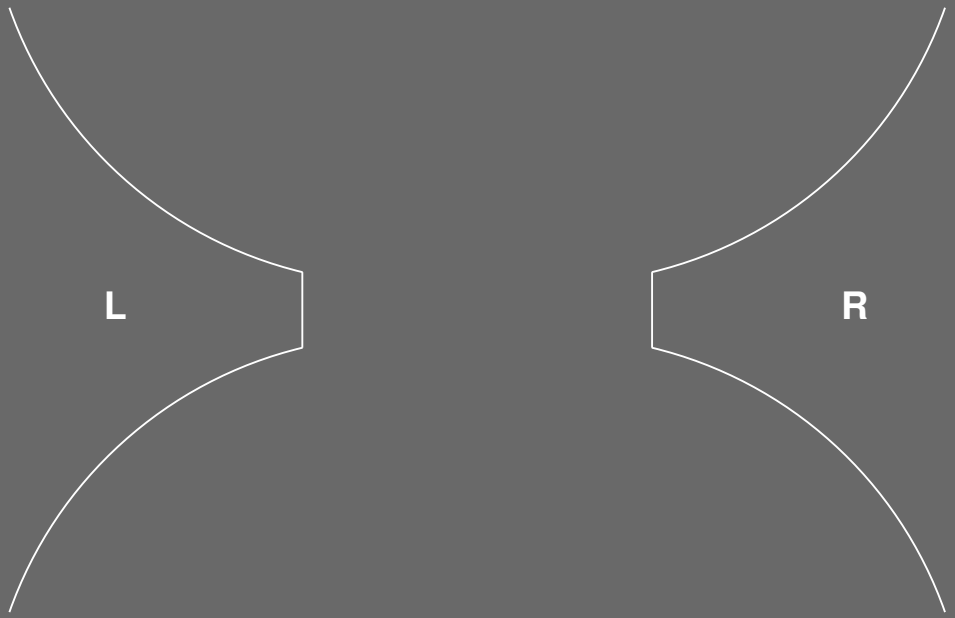
- The accurate calculation of the transmission matrix, t, is key:

$$\mathrm{Tr}\left(tt^\dagger\right) = \mathrm{Tr}\left[\Gamma_L G^r \Gamma_R G^a\right], \tag{2}$$

- Here $\Gamma_{R(L)}$ is the coupling to the right (left) lead and $G^{r(a)}$ is the retarded (advanced) Green's function for the device.

Introduction(3): Previous Work

- Scattering solutions (e.g. Lang, Todorov) are well known
- In effect, we start with *just* the electrodes



- This gives us scattering states ψ_L and ψ_R

Introduction(4): Previous Work

- The connection to the device is given by Lippmann-Schwinger equation:

$$\psi'_L = \psi_L + G \cdot V^{device} \cdot \psi'_L \tag{3}$$

- Here, G is the Green's function for the electrodes
- V^{device} is the *perturbing* potential of the device
- We can achieve the same result with non-equilibrium Green's functions and self-energies

$$G_D = (\epsilon - H_D - \Sigma_L - \Sigma_R)^{-1} \tag{4}$$

- Here the self-energy, Σ_L, is given by $\Sigma_L = h^{\dagger}_{LD} g_L h_{LD}$
- The coupling to leads is $\Gamma_L = i\left[\Sigma_L(\epsilon) - \Sigma_L(\epsilon)^{\dagger}\right]$

Introduction(5): Previous Work

- There are many different implementations:
  - Tight-binding
  - Pure zero-bias scattering
  - DFT (with self-consistency)
  - Full non-equilibrium Green's functions (NEGF)

- Problems or limitations:
  - We can find current-induced forces (Todorov, DiVentra)
  - But solutions are adiabatic
  - Current-induced heating is unsatisfactory
  - Static DFT and excited states ?
  - We are populating empty states for current

Formalism: Time Dependence

- By shifting to the time domain, we find two key advantages:
  - Time-dependent DFT will give us a solid foundation for excited states and transients
  - We can introduce non-adiabatic terms to model current-induced heating of ions
- We consider the simplest possible approach to modelling non-adiabatic effects (Ehrenfest approximation)
- Ions move along unique classical trajectories (no ionic quantum motion; phonons are *classical*)
- Also known as semi-classical approximation

Formalism: Density Matrices

- We present a TB implementation using density matrices
- TB is the simplest quantitative quantum mechanical technique
- Why use the density matrix ?
  - Compact description of electrons
  - Useful in static calculations (link to DFT code)
  - Simplicity of forces
- However, they must be *finite ranged* for practical calculations
- Key problem: Can't use Lippmann-Schwinger equation (link to semi-infinite leads)
- How do we model the environment ?

Formalism: Device and Environment

- System modelled consists of device (system of interest) and environment or leads (open boundaries)
- Environment defined by fixed Hamiltonian and atoms
- Equations of motion for density matrix and ions:

$$\frac{\partial \hat{\rho}}{\partial t} = \frac{1}{\mathrm{i}\hbar}\left[\hat{H}, \hat{\rho}\right] \tag{5}$$

$$M_I \frac{\mathrm{d}^2 \vec{R}_I}{\mathrm{d}t^2} = -\vec{\nabla}_I V_{II} - Tr\{\hat{\rho}\vec{\nabla}_I \hat{H}\} \tag{6}$$

- We will integrate these numerically and simultaneously

Formalism: Device and Environment(2)

- We separate our system into device (D) and environment (E) (or leads)
- We can write:

$$\mathrm{i}\hbar\frac{\partial\hat{\rho}_D}{\partial t} = [\hat{H}_D,\hat{\rho}_D] + (\hat{H}_{DE}\hat{\rho}_{ED} - \hat{\rho}_{DE}\hat{H}_{ED}) \quad (7)$$

$$\mathrm{i}\hbar\frac{\partial\hat{\rho}_{DE}}{\partial t} = \hat{H}_D\hat{\rho}_{DE} - \hat{\rho}_D\hat{H}_{DE} + \hat{H}_{DE}\hat{\rho}_E - \hat{\rho}_{DE}\hat{H}_E \quad (8)$$

$$\mathrm{i}\hbar\frac{\partial\hat{\rho}_E}{\partial t} = [\hat{H}_E,\hat{\rho}_E] + (\hat{H}_{ED}\hat{\rho}_{DE} - \hat{\rho}_{ED}\hat{H}_{DE}) \quad (9)$$

$$- \quad 2\mathrm{i}\hbar\Gamma(\hat{\rho}_E - \hat{\rho}_{ref}). \quad (10)$$

- The final term ($-\mathrm{i}\hbar\Gamma(\hat{\rho}_E - \hat{\rho}_{ref})$) is a damping term
- It represents inelastic scattering that takes the system back to ρ_{ref}

Formalism: Environment

- For the environment, we can find a closed form
- We define *driving terms:*

$$\mathrm{i}\hbar\hat{G}_E = (\hat{H}_{ED}\hat{\rho}_{DE} - \hat{\rho}_{ED}\hat{H}_{DE}) \quad (11)$$

$$0 = [\hat{H}_E, \hat{\rho}_E(0)] + \mathrm{i}\hbar\hat{G}_E^{(0)} - 2\mathrm{i}\hbar\Gamma(\hat{\rho}_E(0) - \hat{\rho}_{ref}) \quad (12)$$

- Using the interaction picture, and assuming *time independent* $\hat{H}$, we find:

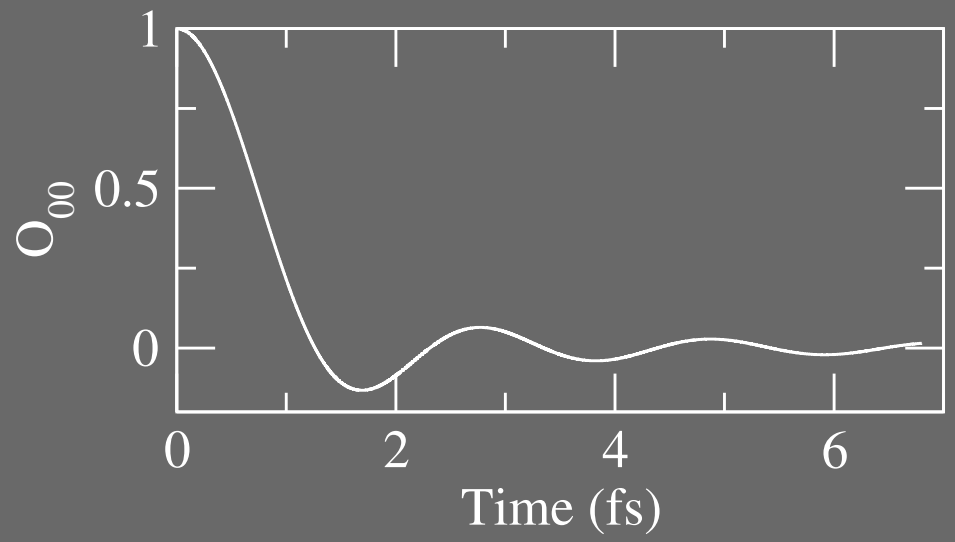
$$\hat{\rho}_E(t) = \hat{\rho}_E(0) + \int_0^t \mathrm{d}x\, \hat{O}(x)\left(\hat{G}_E(t-x) - \hat{G}_E^{(0)}\right)\hat{O}^\dagger(x) \quad (13)$$

where $\hat{O}(t) = \mathrm{e}^{-\Gamma t}\mathrm{e}^{\hat{H}_E t/\mathrm{i}\hbar}$

- Note that eq. 13 is non-local in time (*non-Markovian*)

Formalism: Environment(2)

- Find O matrix using Green's functions
- How does the O matrix behave over time ?



- It is short-ranged: we can truncate in time
- This will also result in spatial truncation: finite propagation

Formalism: Device

- The device is easier: we allow $\hat{H}$ to vary, and simply integrate numerically:

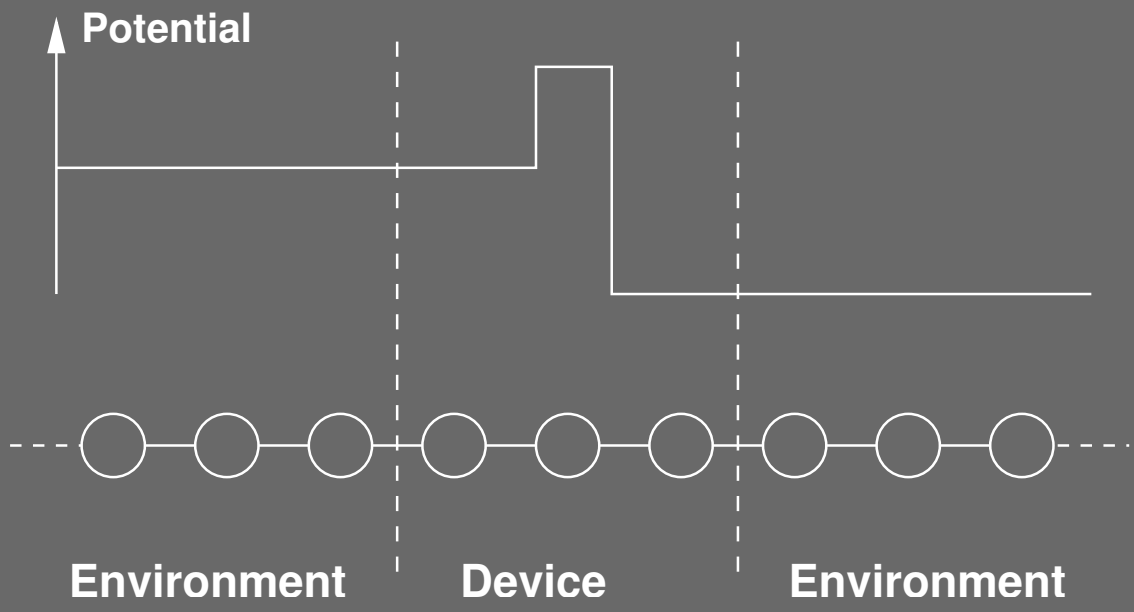
$$\frac{\partial \hat{\rho}_D}{\partial t} = \frac{1}{\mathrm{i}\hbar}\left[\hat{H}_D, \hat{\rho}_D\right] + \frac{1}{\mathrm{i}\hbar}(\hat{H}_{DE}\hat{\rho}_{ED} - \hat{\rho}_{DE}\hat{H}_{ED}) \quad (14)$$

$$\rho_D(t+\Delta t) \approx \rho_D(t-\Delta t) + 2\Delta t \rho'_D(t) \quad (15)$$

- **BUT** we have to assume that the matrix $\hat{\rho}_{DE}$ is **short-ranged**
- This is reasonable:
  - We know that, with scattering, ρ is local
  - We assert that the environment is **large**: the effect of the device is small
- We can also view $\frac{1}{\mathrm{i}\hbar}(\hat{H}_{DE}\hat{\rho}_{ED} - \hat{\rho}_{DE}\hat{H}_{ED})$ as a driving term resulting from truncation

Results

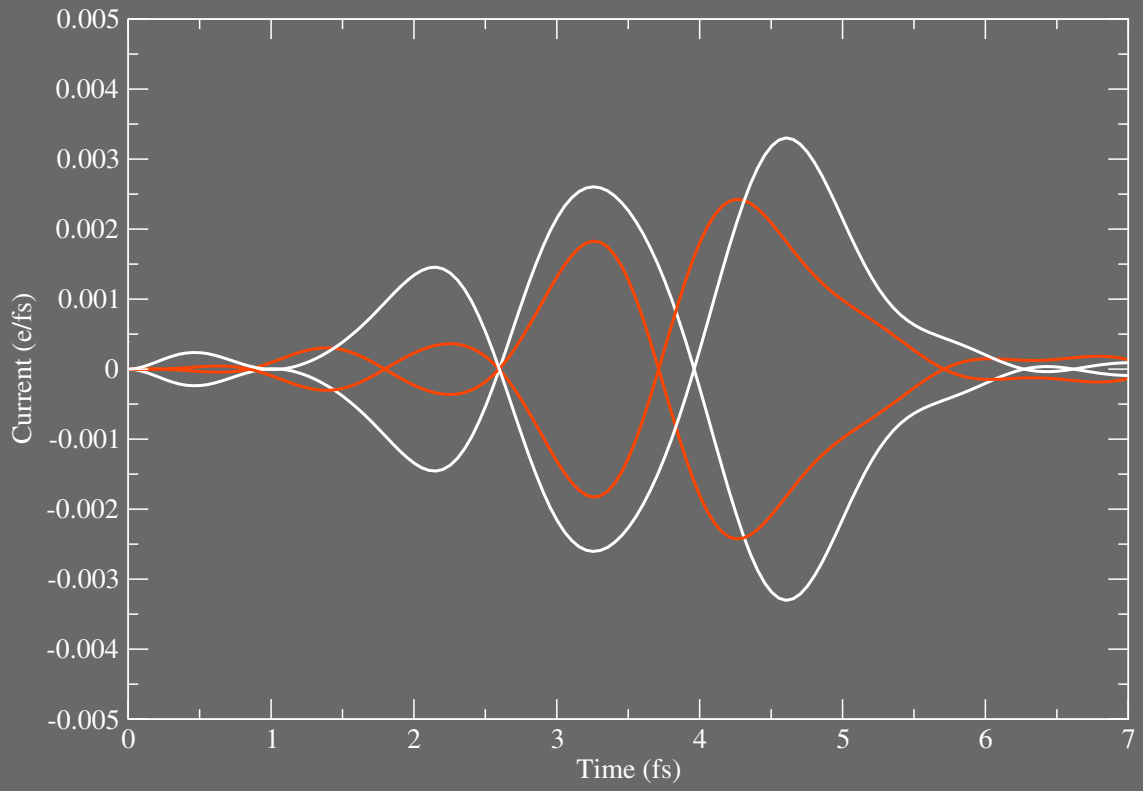
- Simple model: 1D chain of atoms, tight binding, 3 atom device, central atom barrier



- Nearest neighbour hopping: $t_{ii+1} = t_0 f(r_{ii+1})$
- Timestep 2 attos (2×10^{-18}s)

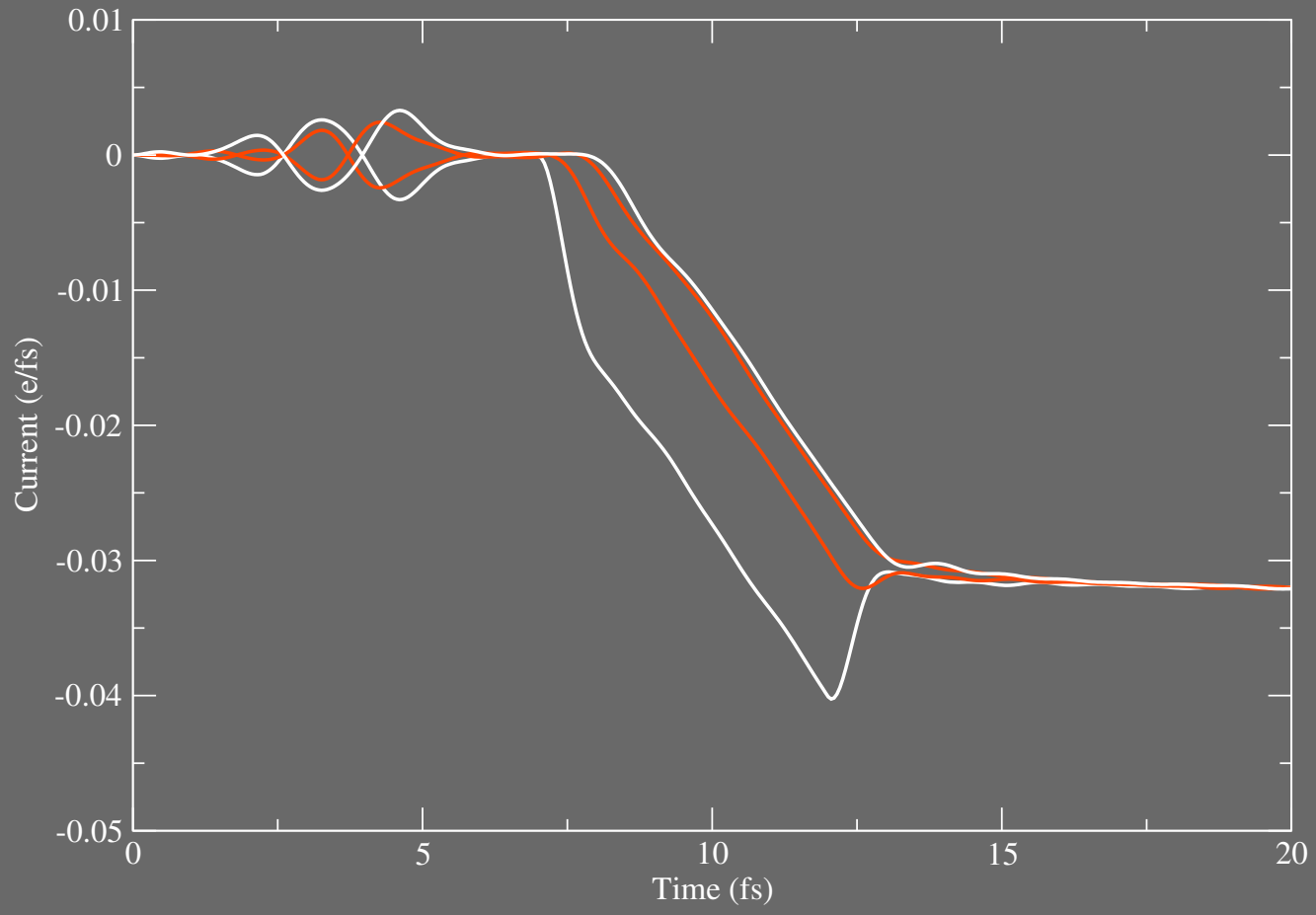
Results: Zero bias

- In TB we can write $I_{ii+1} = H_{ii+1}\mathrm{Im}\left(\rho_{ii+1}\right)$
- Plot current in and out of device (white), in and out of scatterer (orange)
- At zero bias, the current settles to a steady state



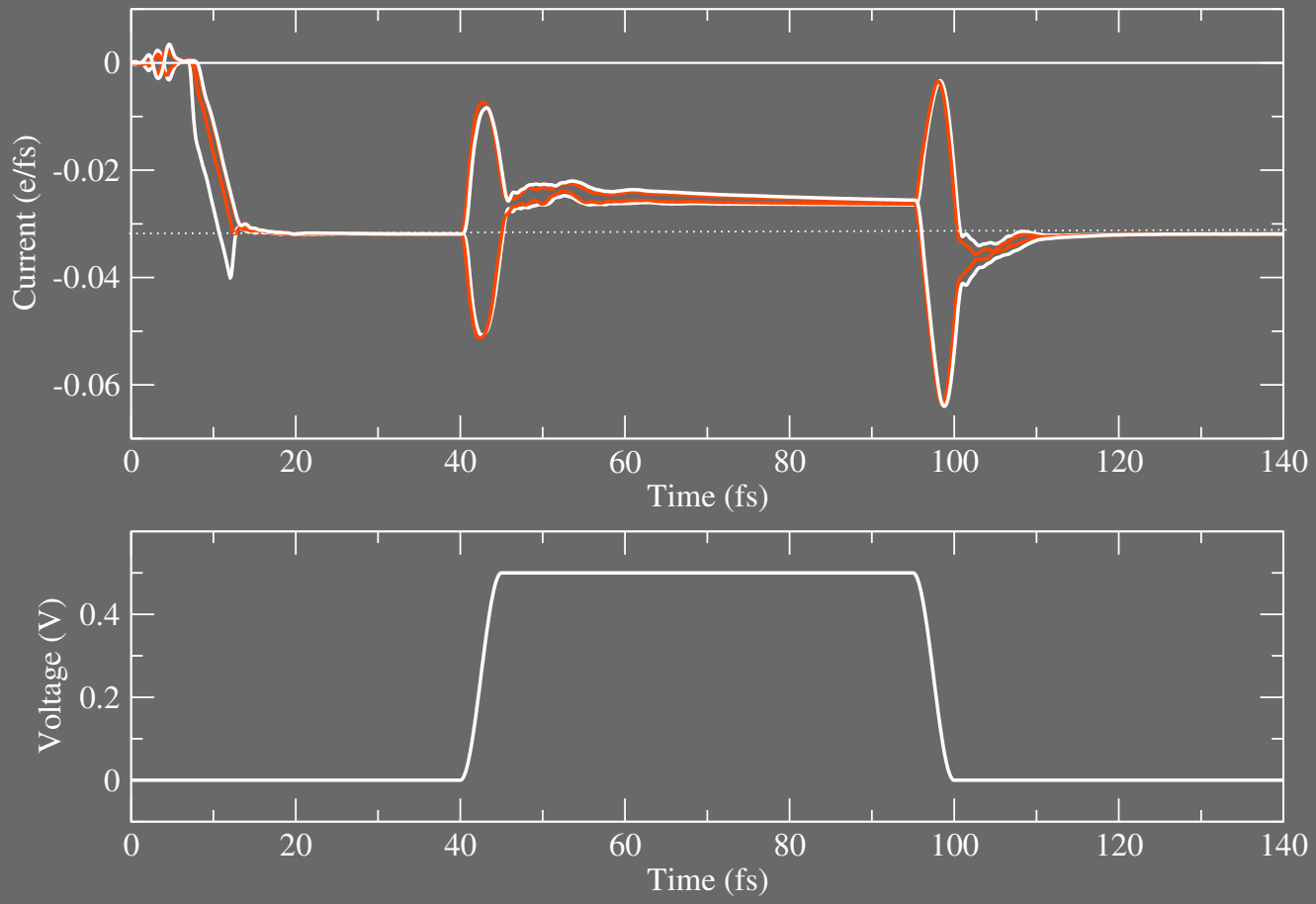
Results: Switching on bias

- We switch on a bias of -0.1V from 7.5 to 12.5 fs



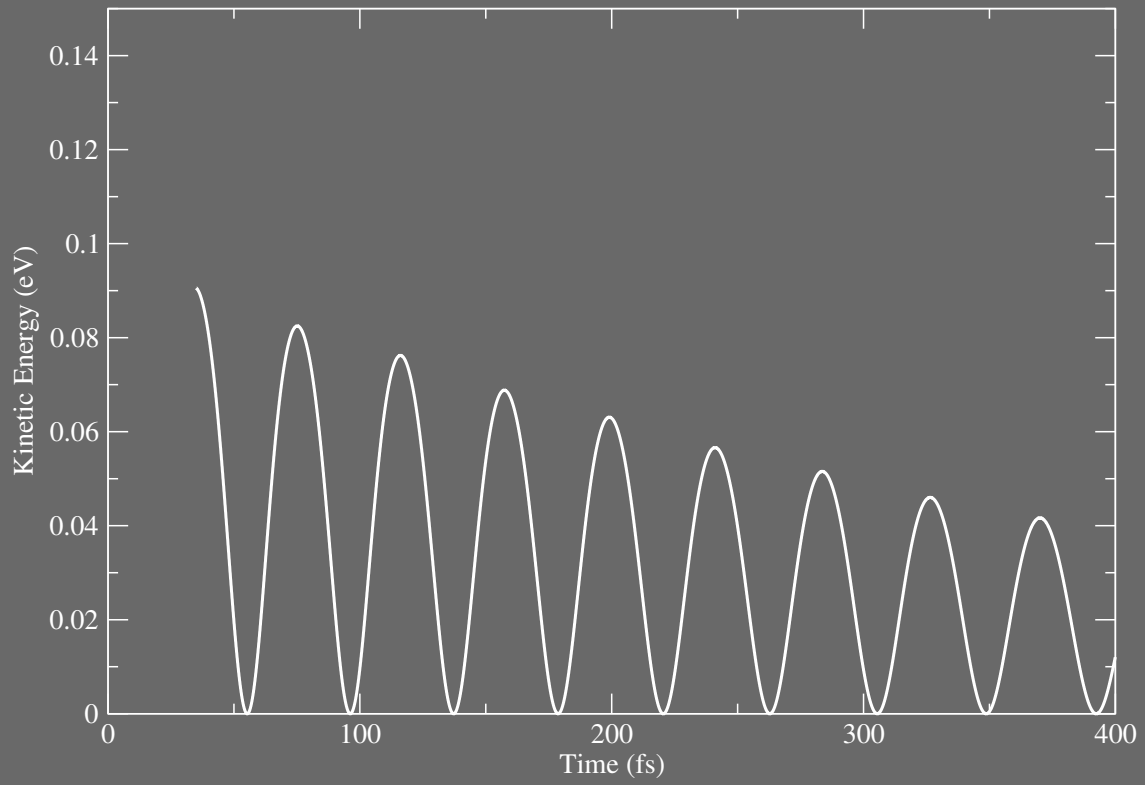
Results: Applying gate voltage

- Apply varying “gate voltage” to central atom



Results: Cooling with small bias

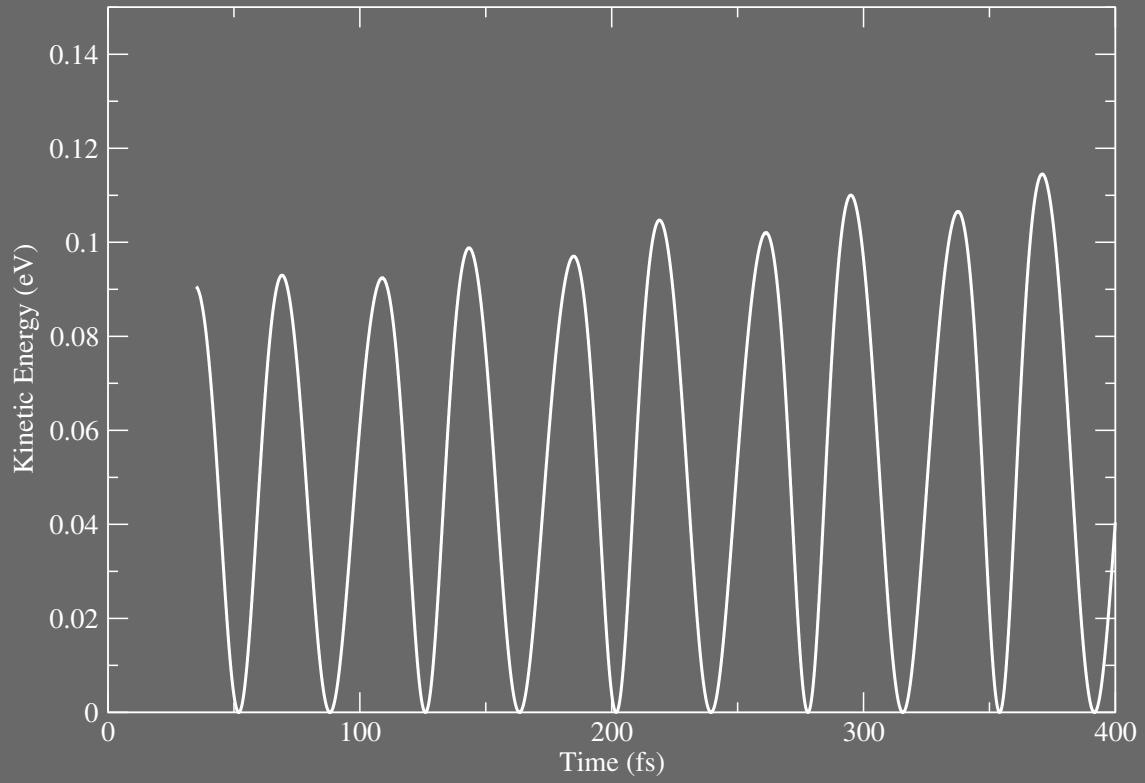
- Allow central atom to move after 35fs with bias of -0.1V: see cooling



- Vibrational frequency has energy ($\hbar\omega$) of 0.055 eV

Results: Heating with large bias

- Bias -1.0V: large heating effect



- Small bias only allows limited range of transitions; large bias allows transitions between a significant range of BO surfaces

Conclusions

- New formalism for time evolution of density matrix
- Requires non-Markovian time evolution
- A steady state can be achieved (with damping)
- “Gate voltage” effects show sensible, physical behaviour
- Heating and cooling are seen
- Intend to look at implementing quantum contributions to ionic motion

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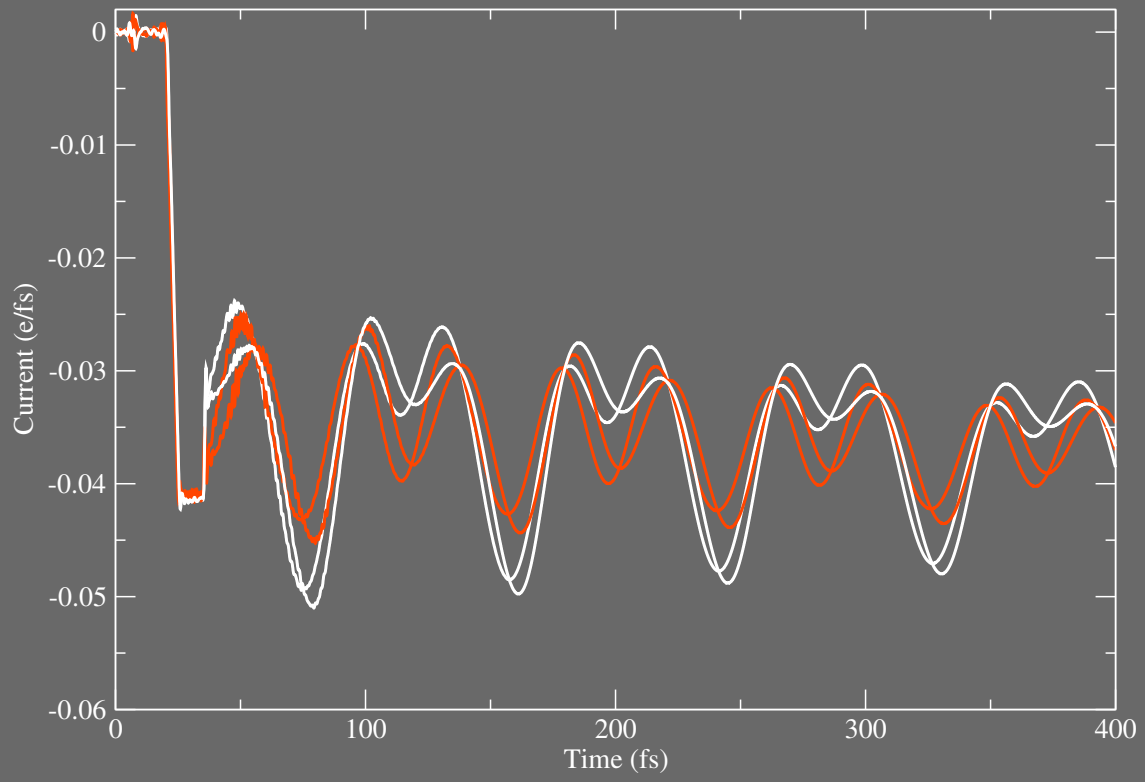
Results: Current during cooling

- Cooling current



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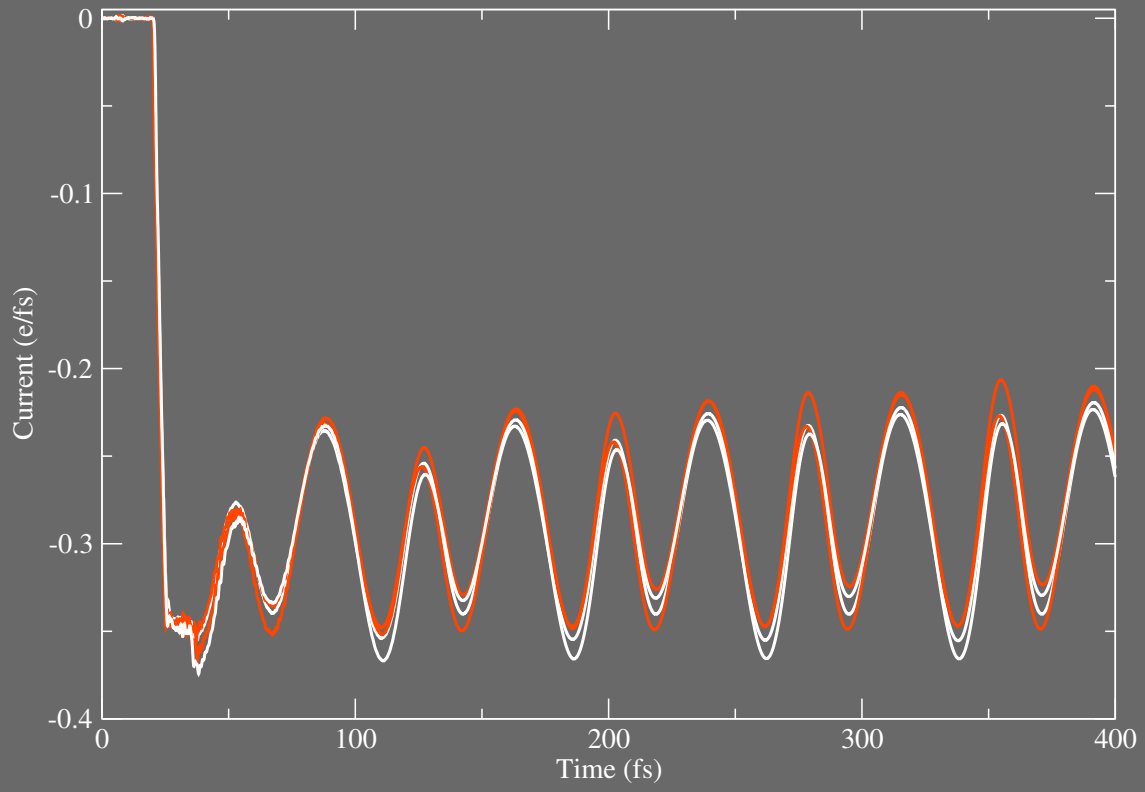
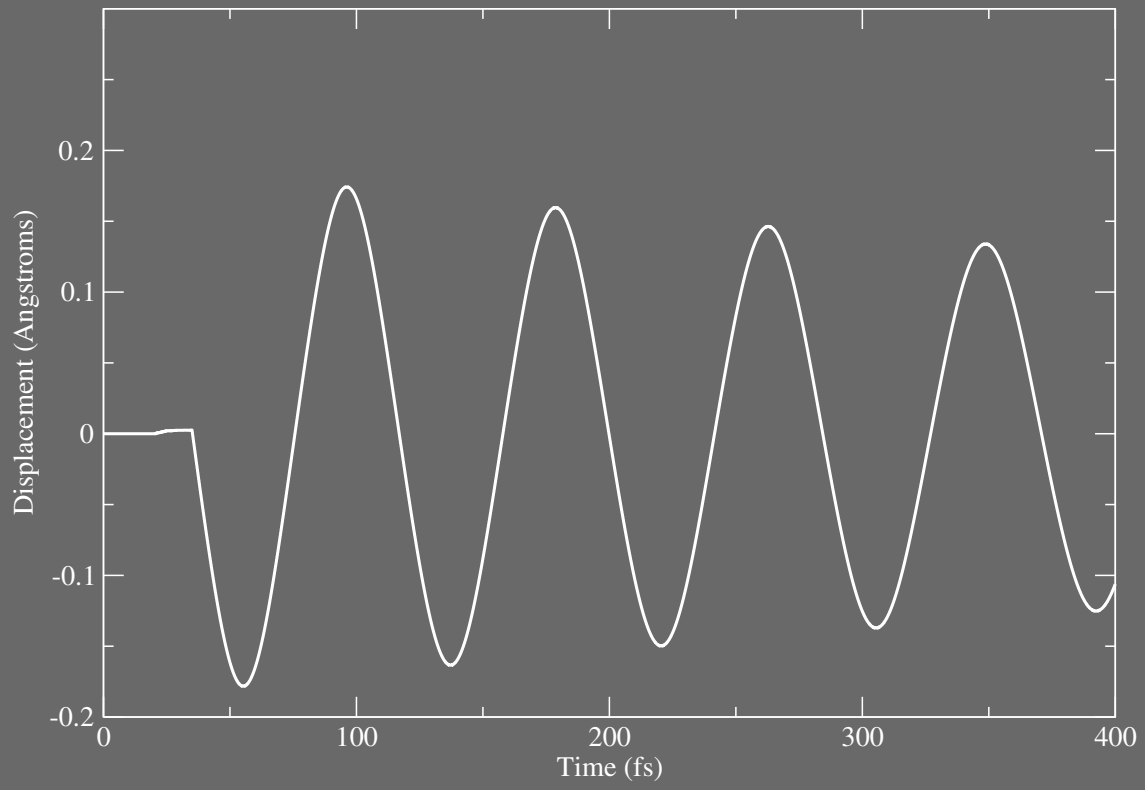


Results: Positions during cooling

- Cooling position (central atom)



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