A model for inelastic transport through atomic surface wires

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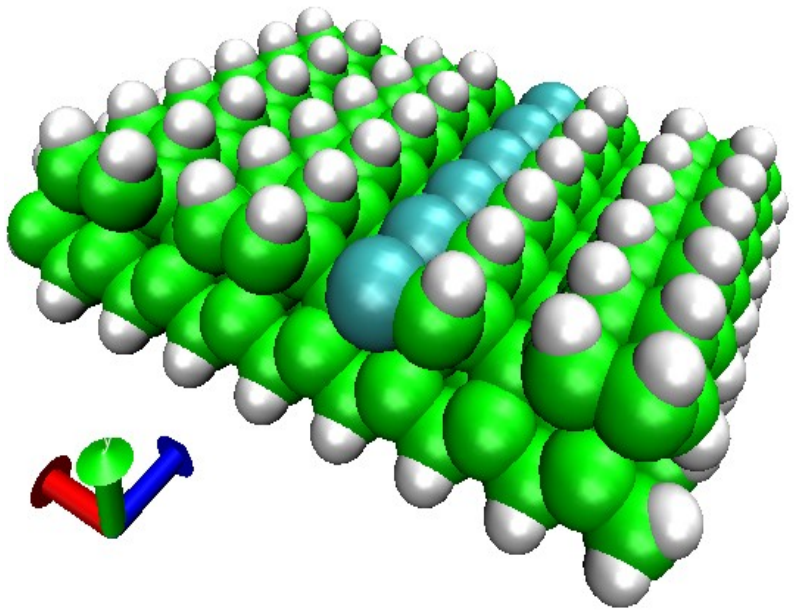
A model for inelastic transport through atomic surface wires

1. Short introduction
2. Random distortion of the wire
3. Mixed quantum/classical dynamics
4. A quantum model for transitions

1. Atomic wires

One dimensional atomic wire : dangling bond wire at the SiH surface

Transport calculation and heating

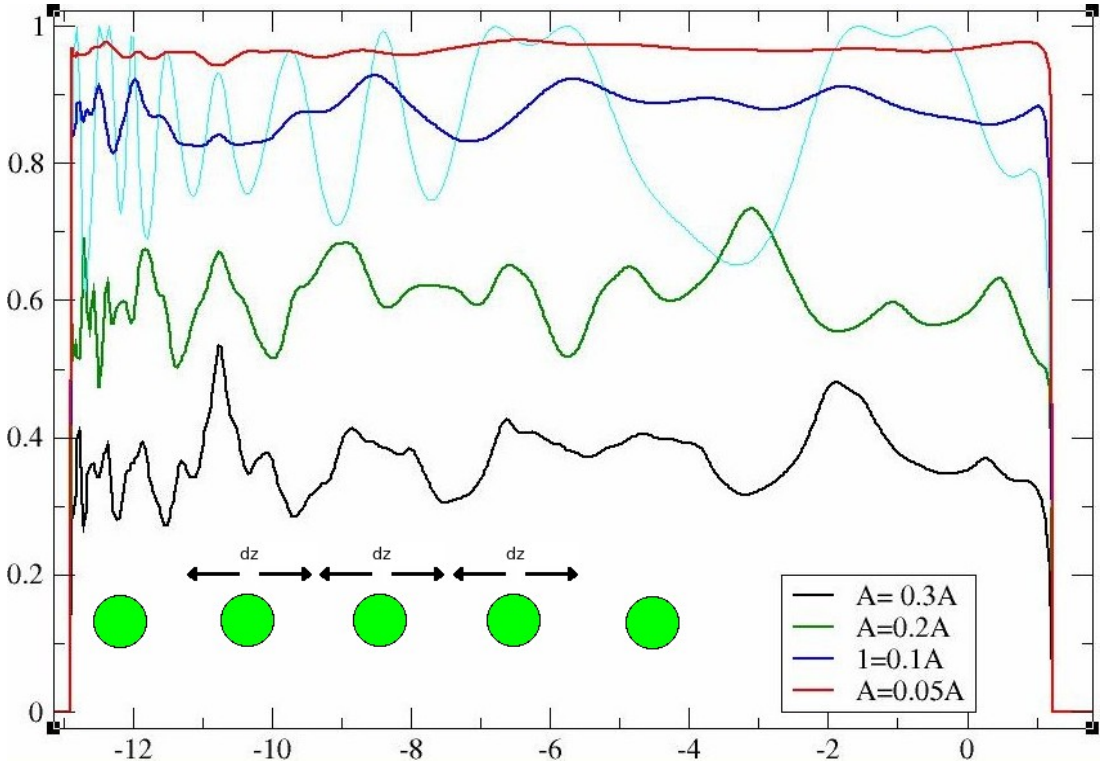


2. Random distortions

Linear atomic gold wire

Displacements are given by a uniform random distribution.

The curves show the dependence of the transmission on the amplitude of the displacements.



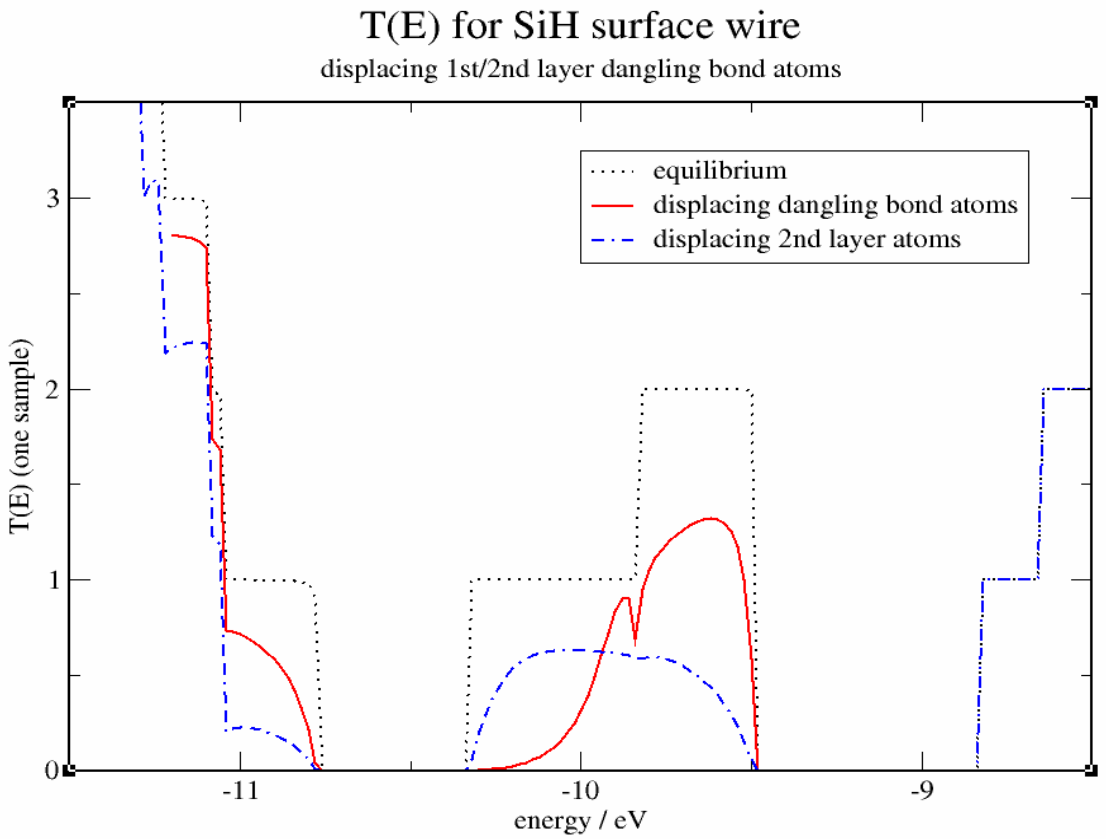
Work of Eeva Niemi in Toulouse, to be published.

2. Random distortions

1D dangling bond wire

Displacements are given by a uniform random distribution.

The curves show the dependence of the transmission on the number of atomic layers displaced. Maximum amplitude is 20% of the Si-Si distance.



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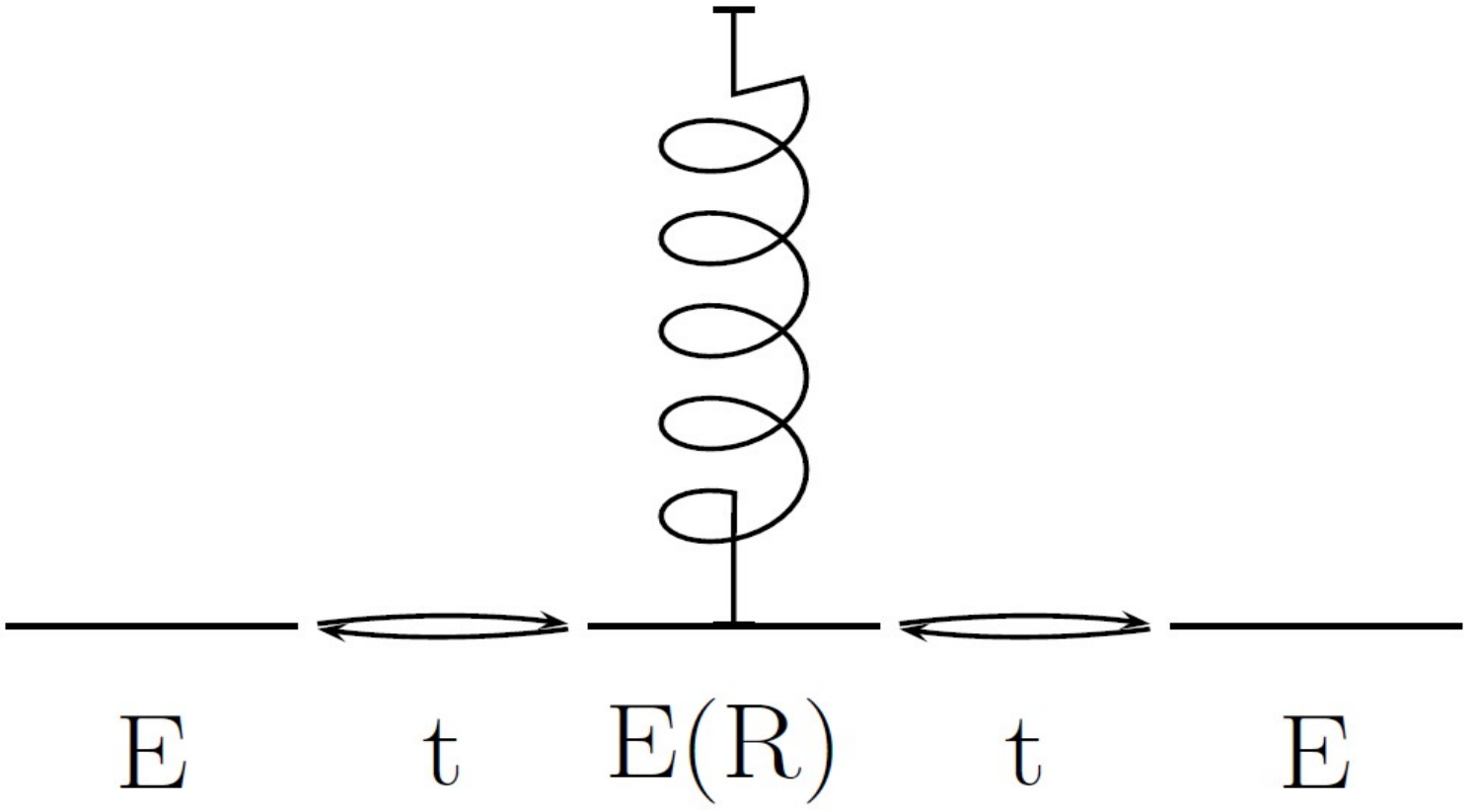
3. Ehrenfest dynamics

The model

$$h = \begin{pmatrix} E & t & 0 \\ t & E & t \\ 0 & t & E \end{pmatrix}$$

3. Ehrenfest dynamics

The model



3. Ehrenfest dynamics

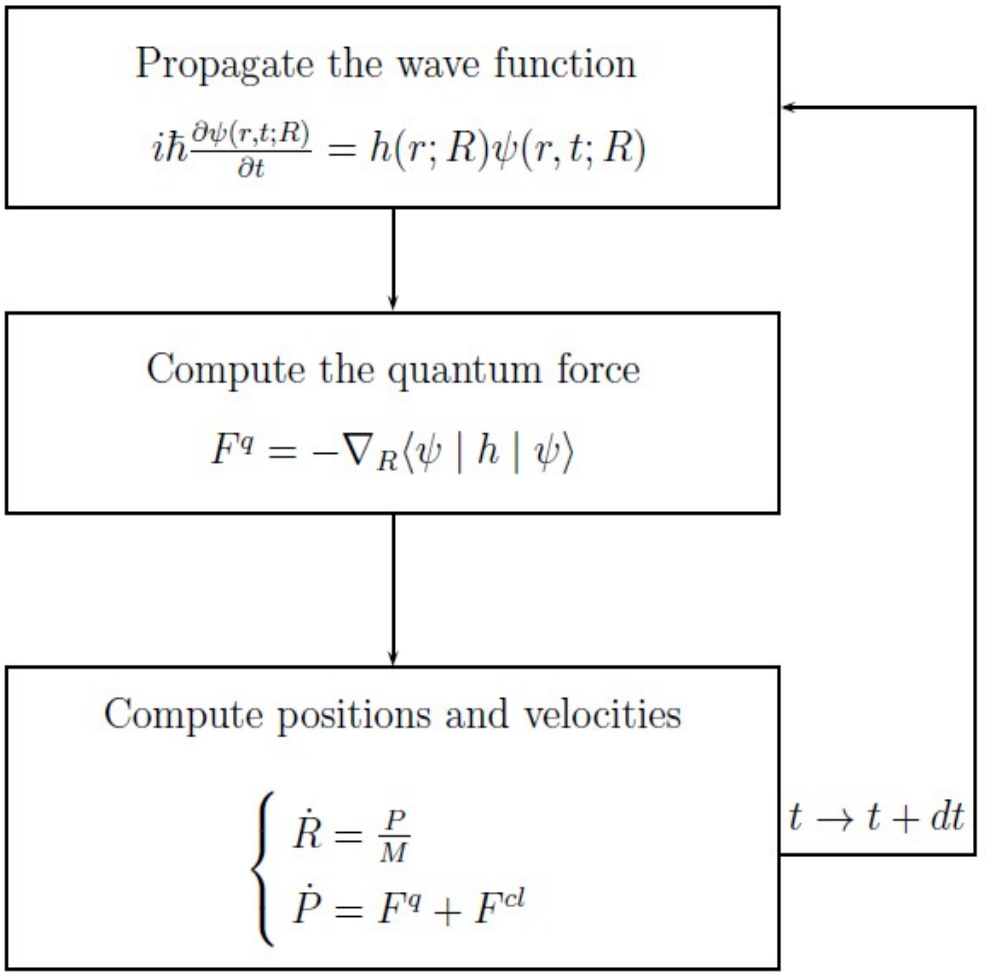
The model

$$h(R) = \begin{pmatrix} E & t & 0 \\ t & E(R) & t \\ 0 & t & E \end{pmatrix} \qquad E(R) = e + \frac{1}{2}K(R - R_{eq})^2$$

- Nuclear coordinate « R » representation, **classical** harmonic oscillator
- Electronic coordinate State representation, explicit **quantum** dynamics

3. Ehrenfest dynamics

The method



3. Explicit dynamics

The method

Mean field theory guaratees the conseravtion of the total energy throughout the propagation

- Electronic part

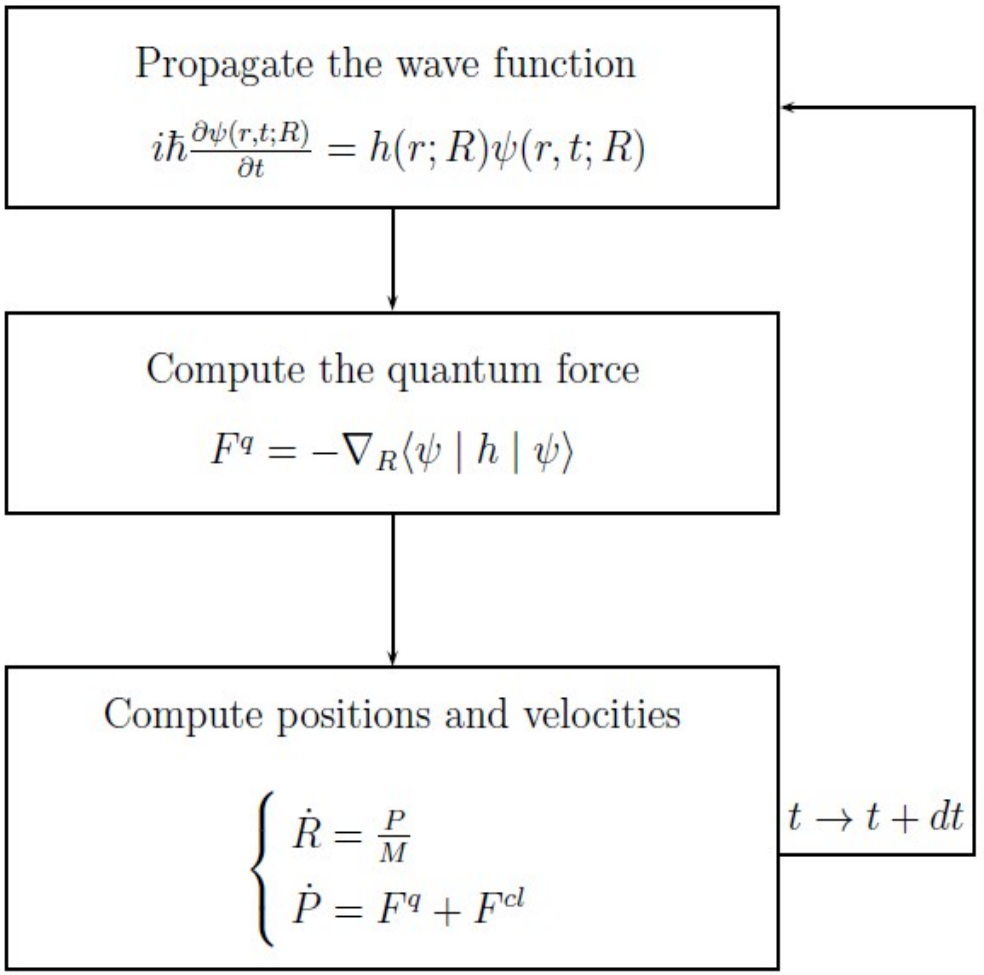
Direct diagonaisation or short iterative Lanczos

- Classical part

Velocity-corrected Verlet

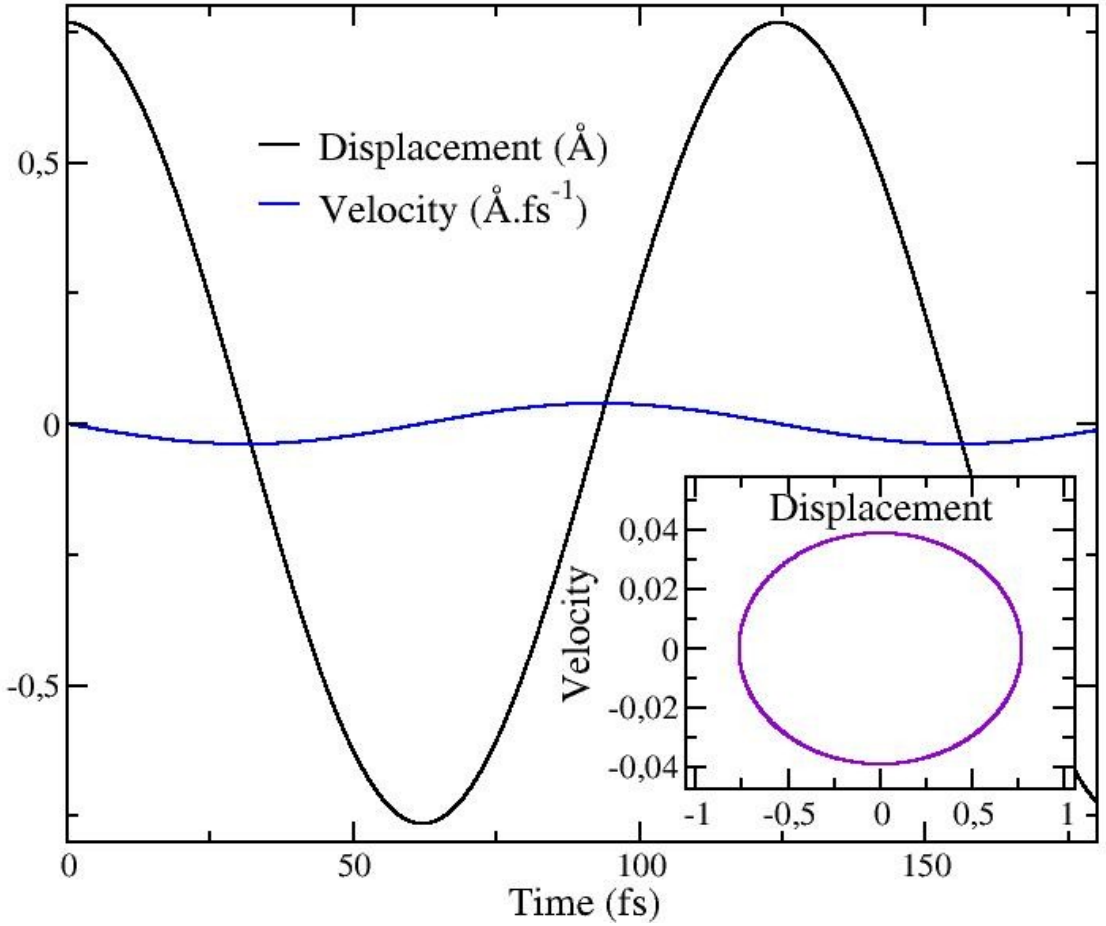
- Force

Finite differences



3. Ehrenfest dynamics

Some results : positions ans velocities



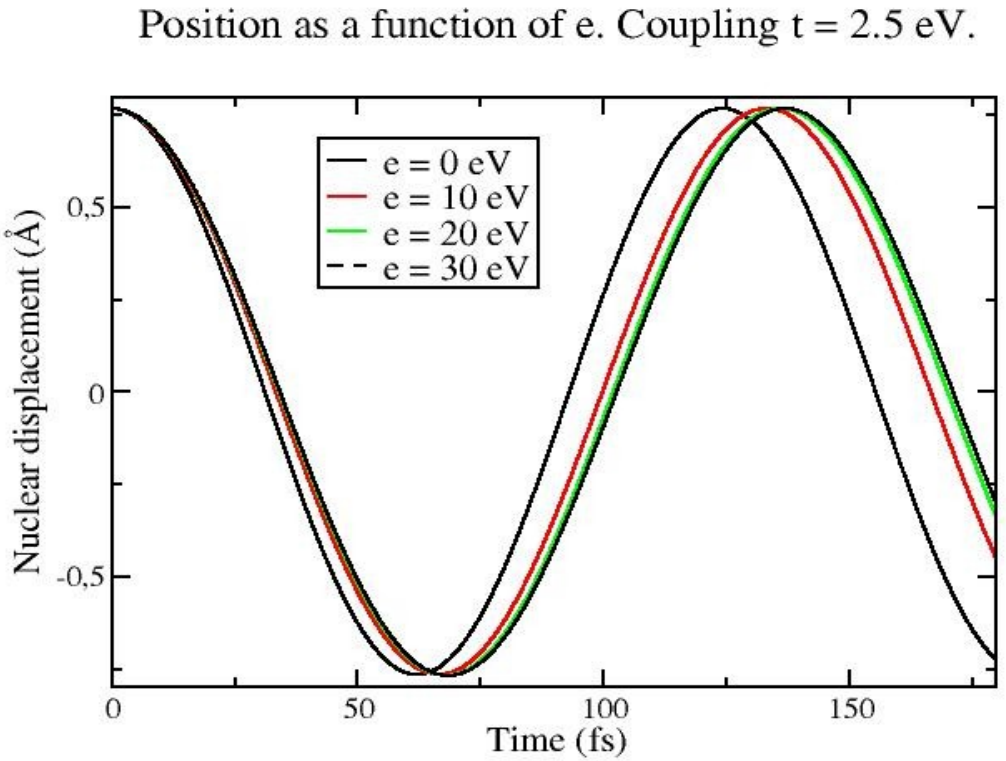
Frequency 30 meV (Barcelona's group result)
Spring constant 6 eV.Å^{-2}
Time step 10^{-3} fs

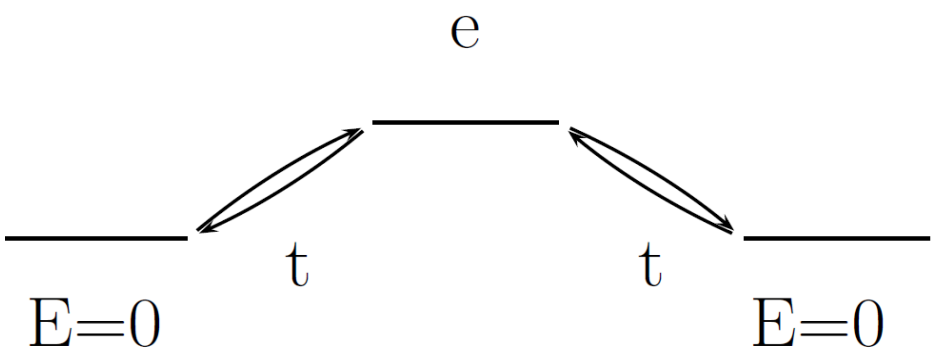
<u>Starting parameters</u>

- Zero velocity
- Displacement : 20% of Si-Si distance along dangling bond rows

3. Ehrenfest dynamics

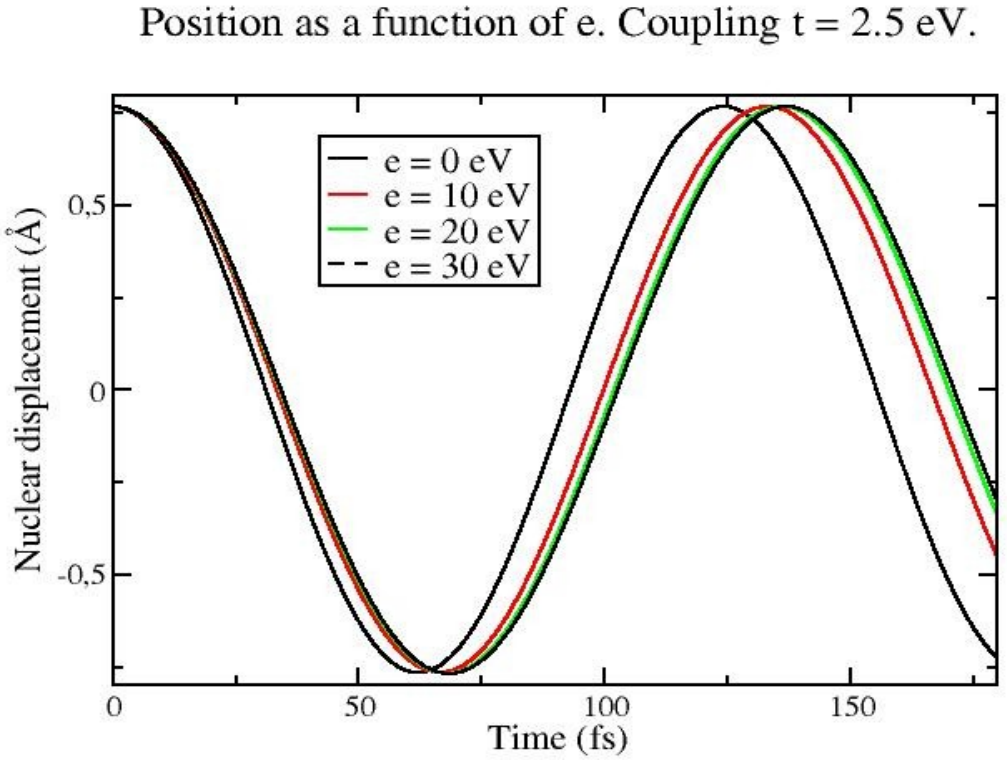
Some results : displacements

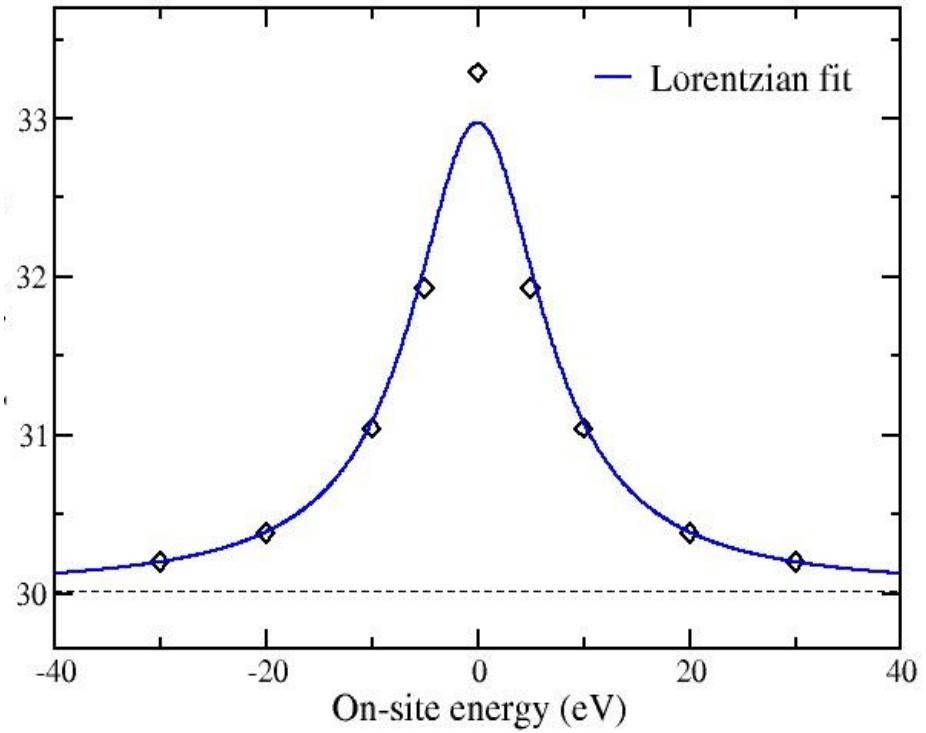




3. Ehrenfest dynamics

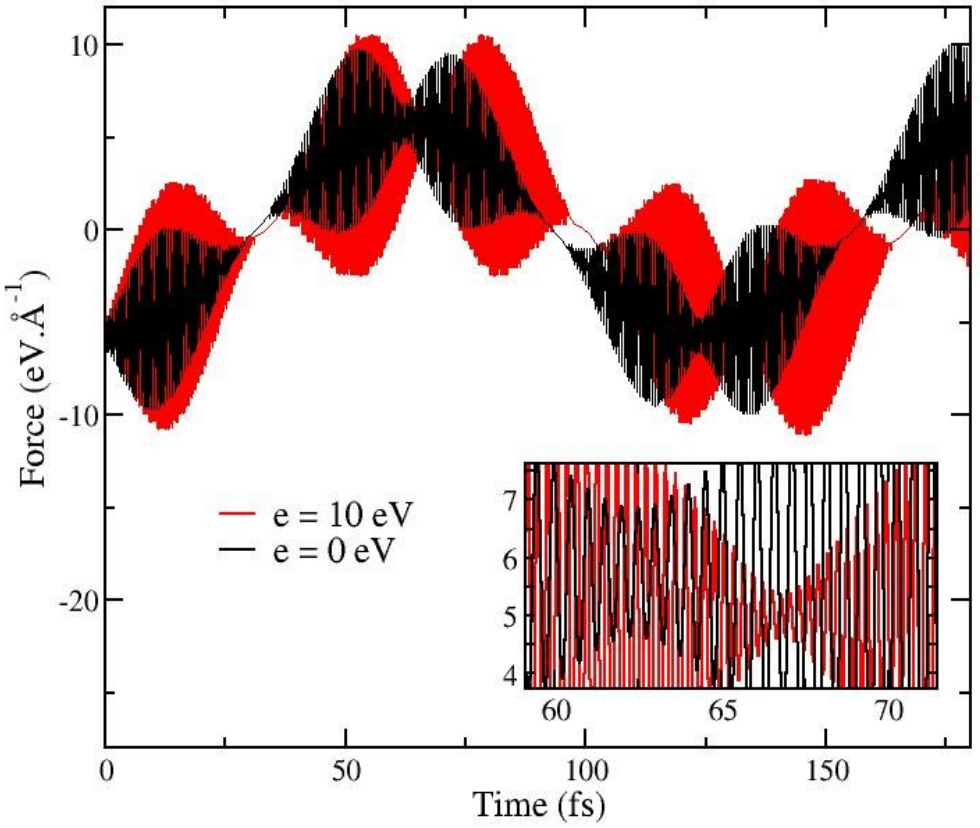
Some results : displacements



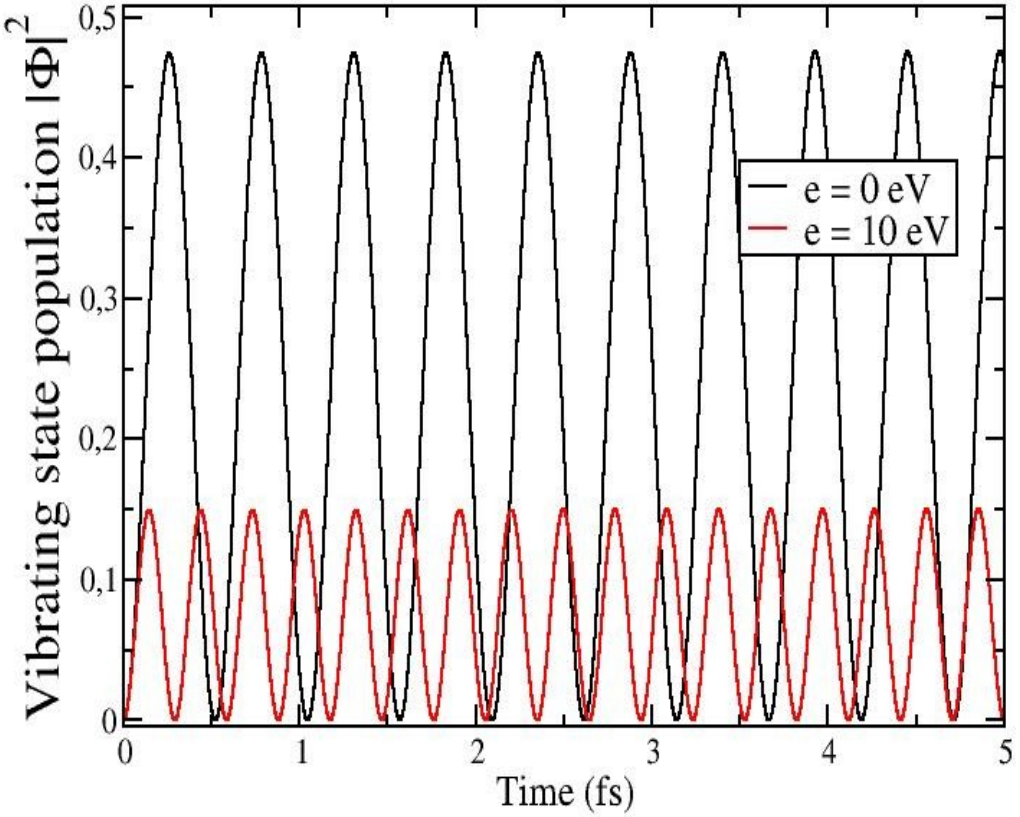


3. Ehrenfest dynamics

Some results : the force

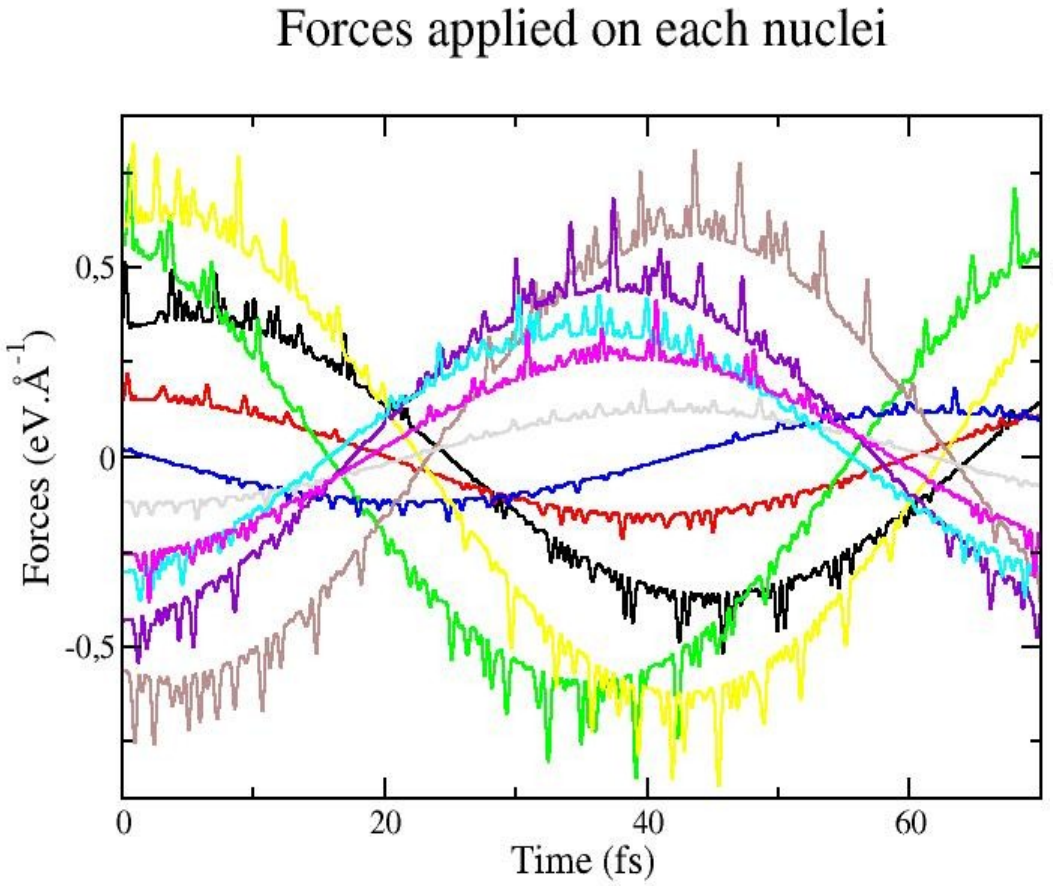


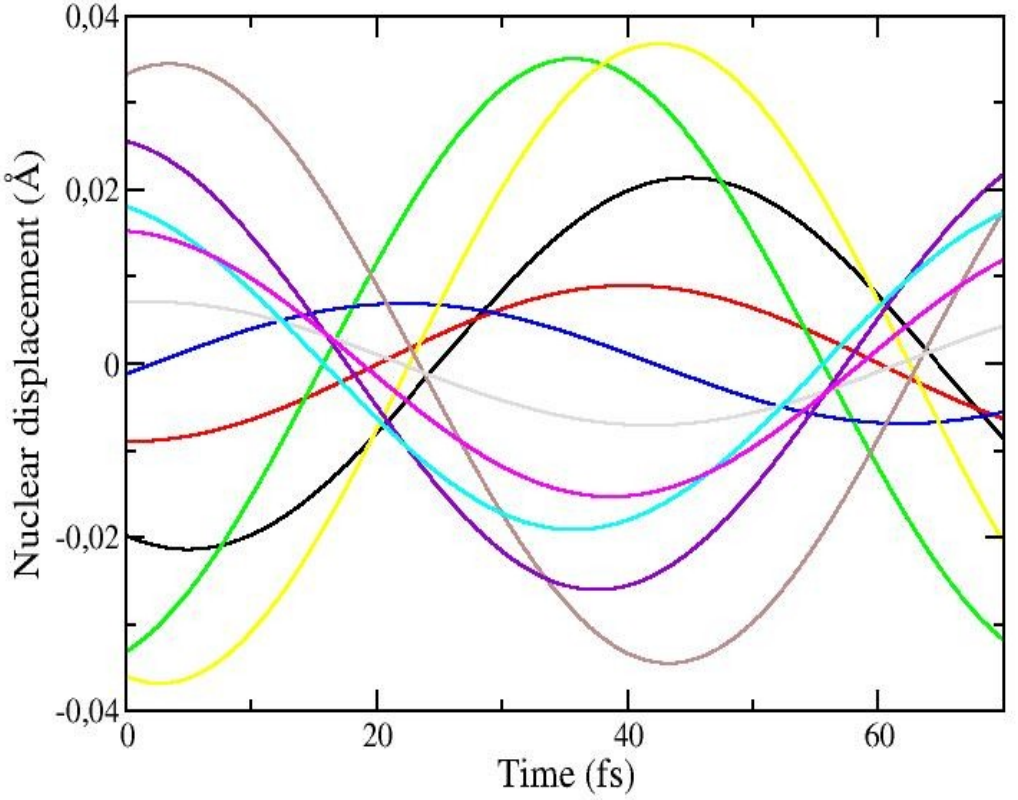
e = 0 or 10 eV with coupling t = 2.5 eV



3. Ehrenfest dynamics

Calculations with 10 vibrating sites





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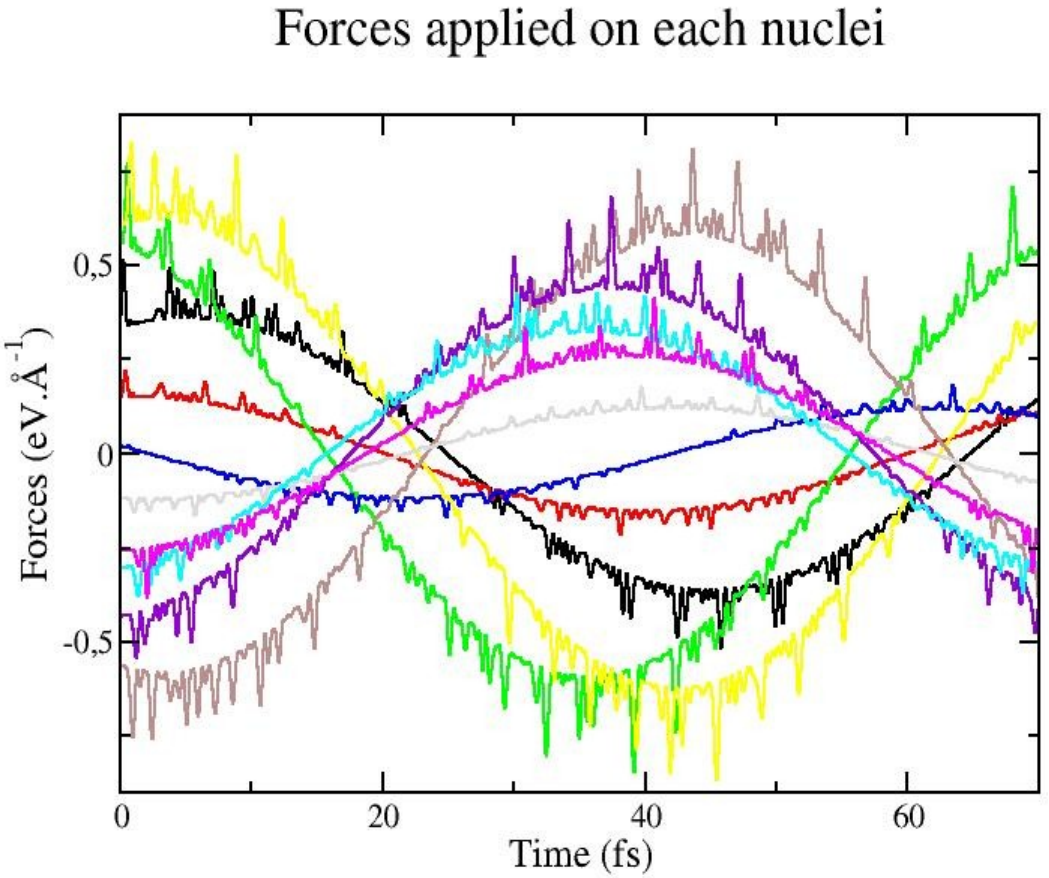

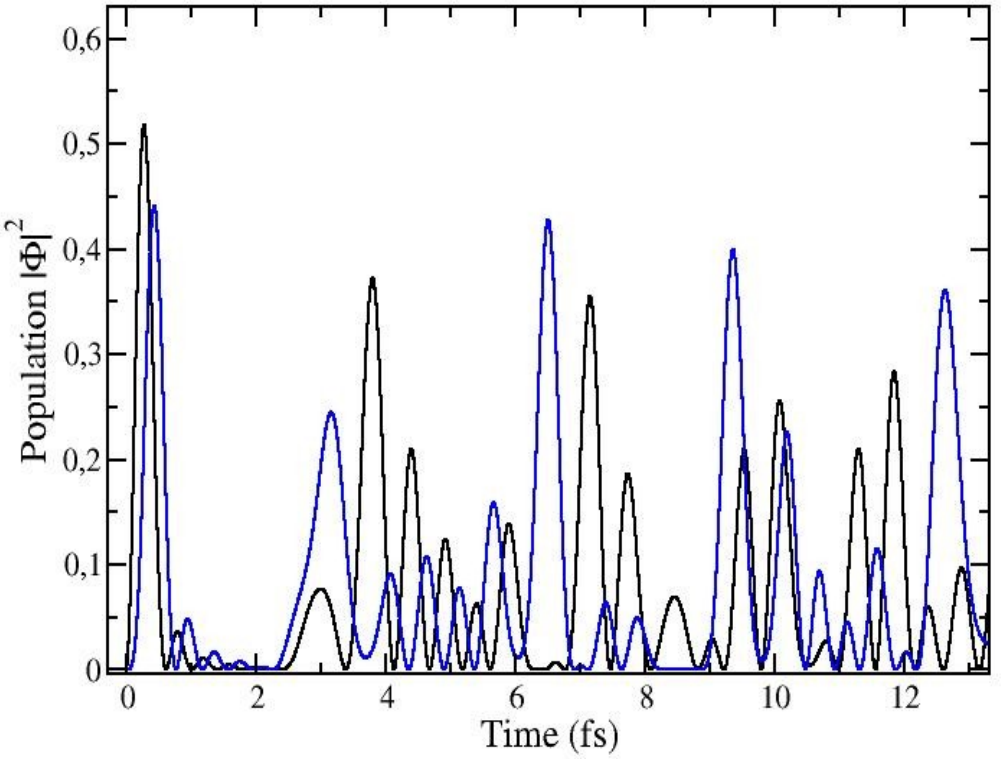

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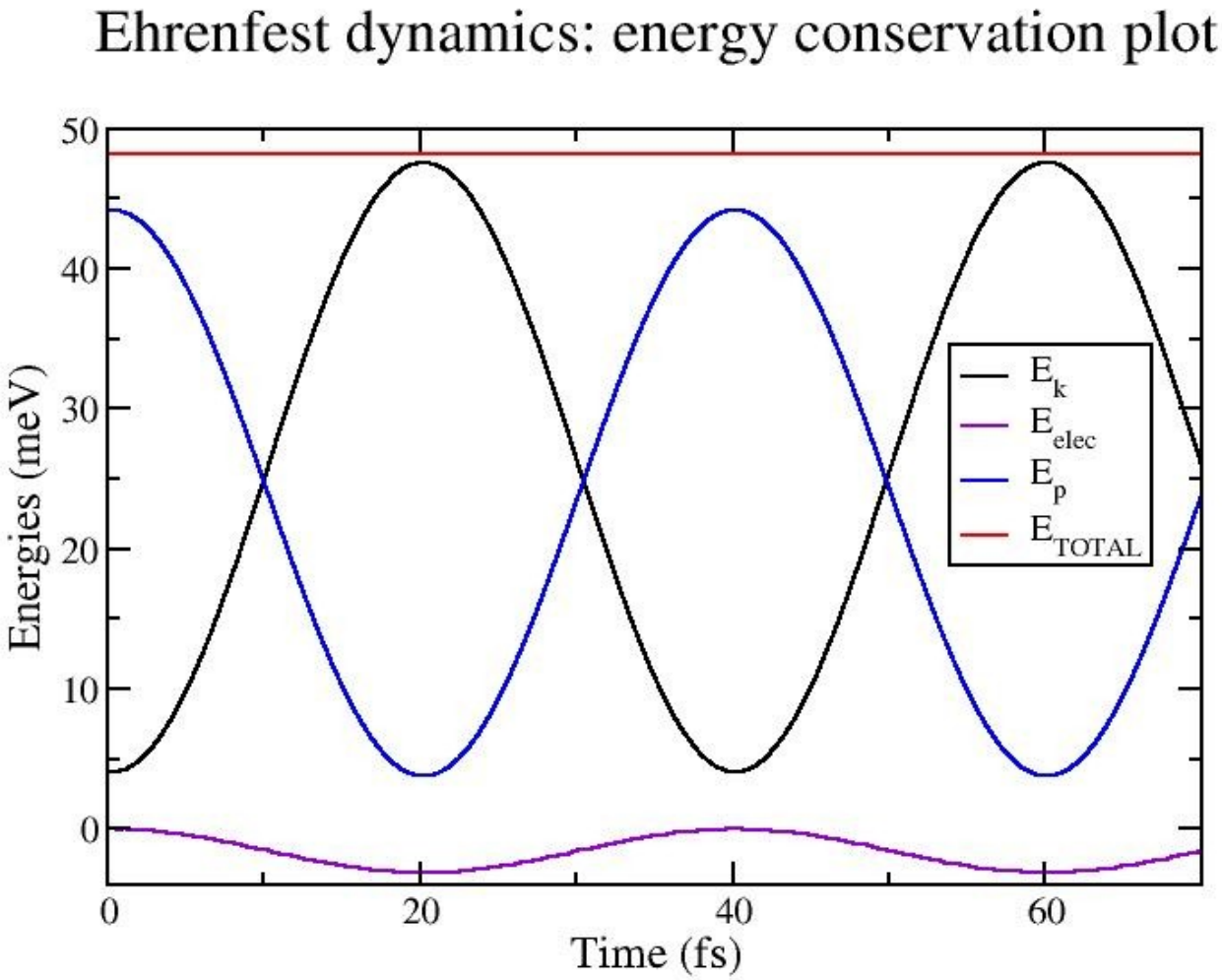
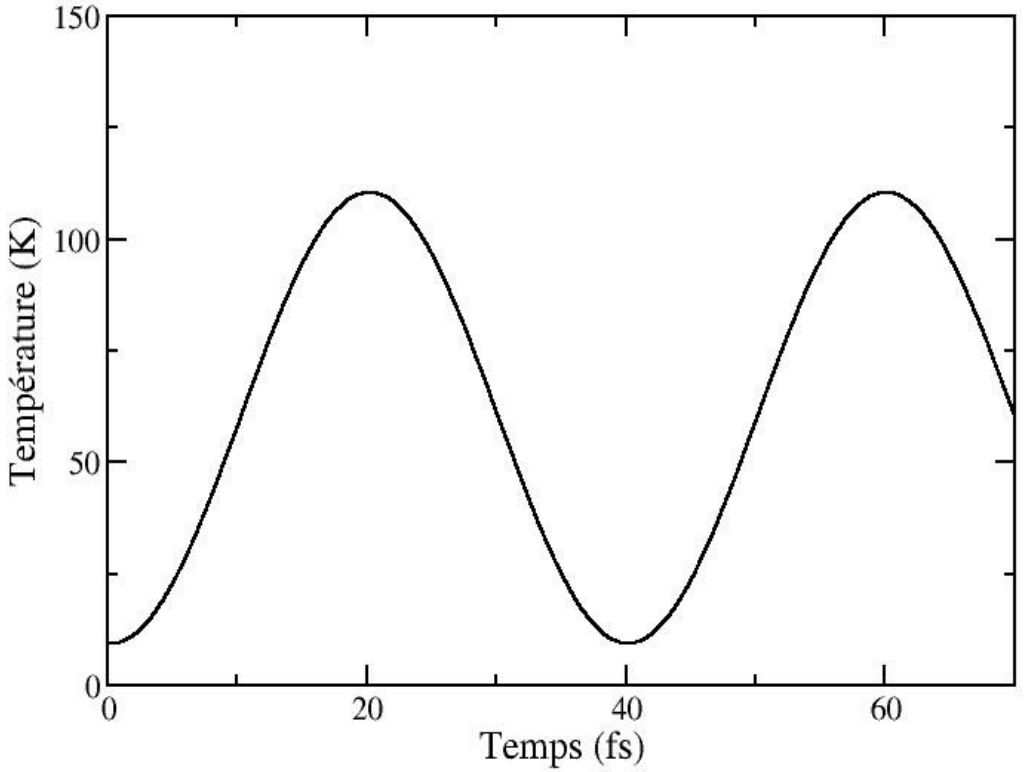
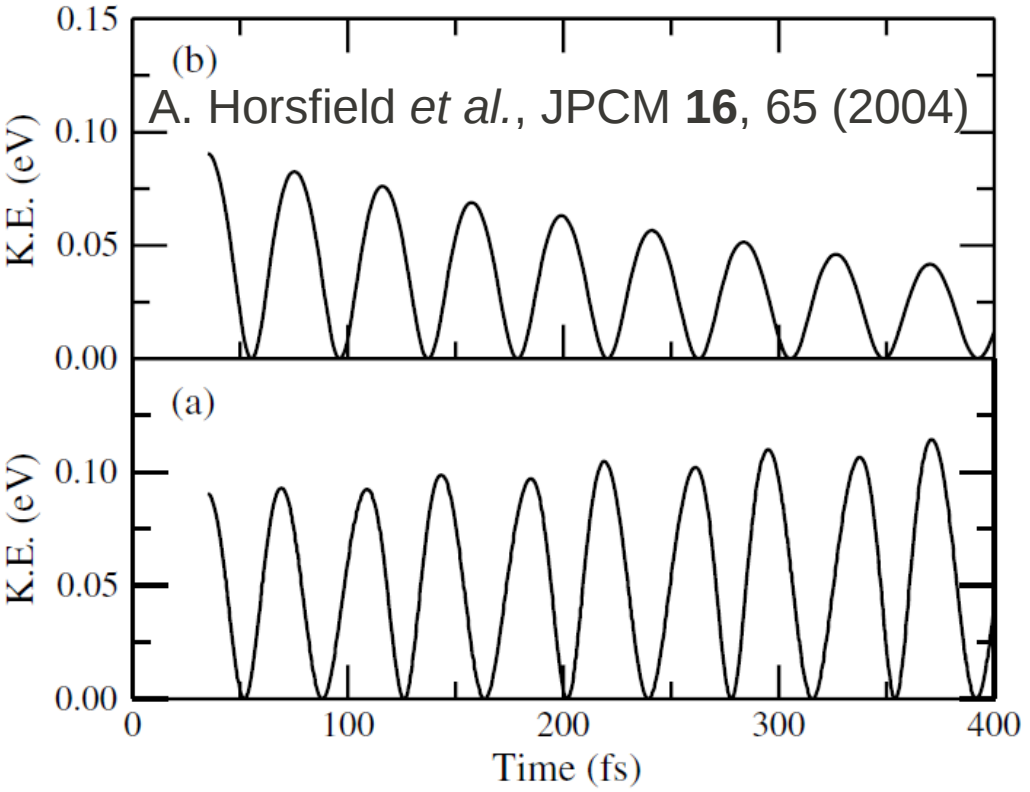


3. Ehrenfest dynamics

Heating : the electrons release energy to the nuclear degrees of freedom irreversibly

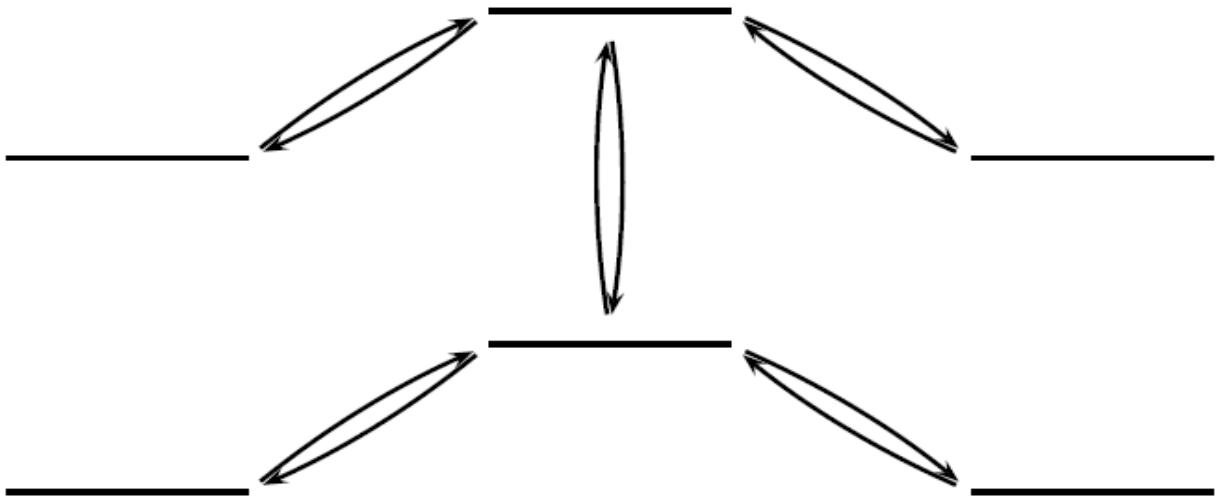




A. Horsfield *et al.*, JPCM **16**, 65 (2004)

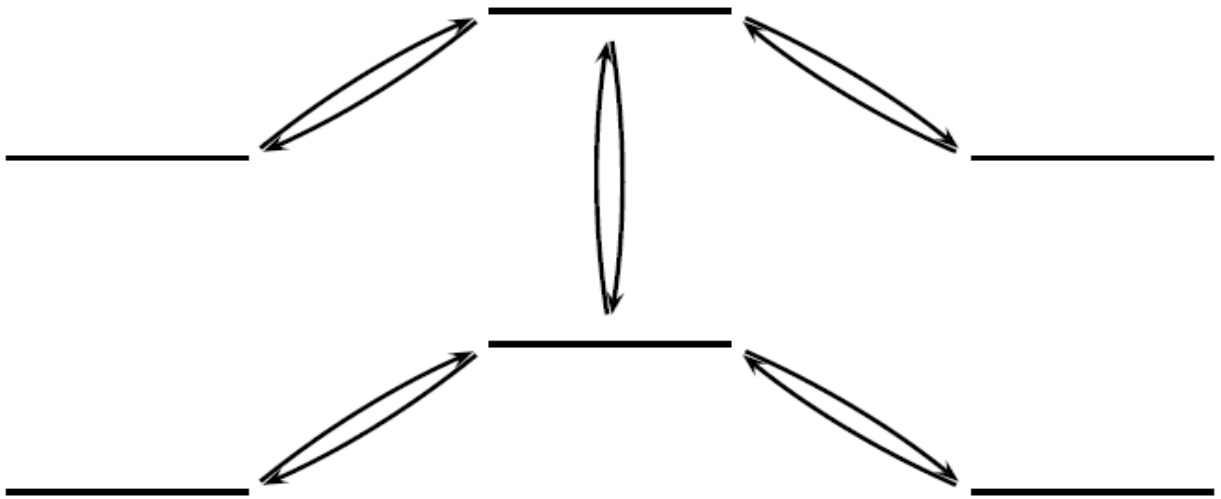
4- A quantum model

Motivation : Are there any rules for vibrational transitions?



4- A quantum model

6 states : 3 electronic states coupled to a vibration (n=0, 1)



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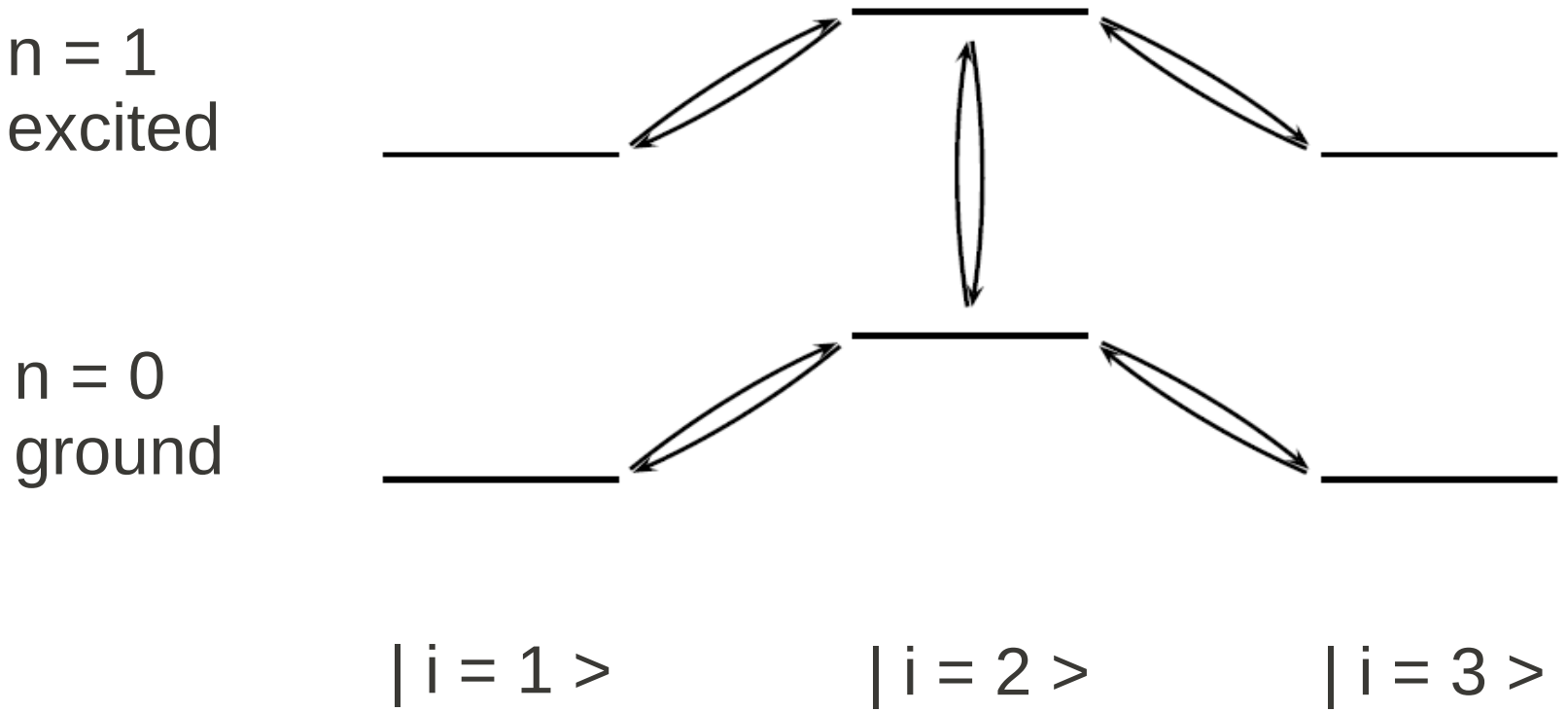
$$| i > \otimes | n >$$



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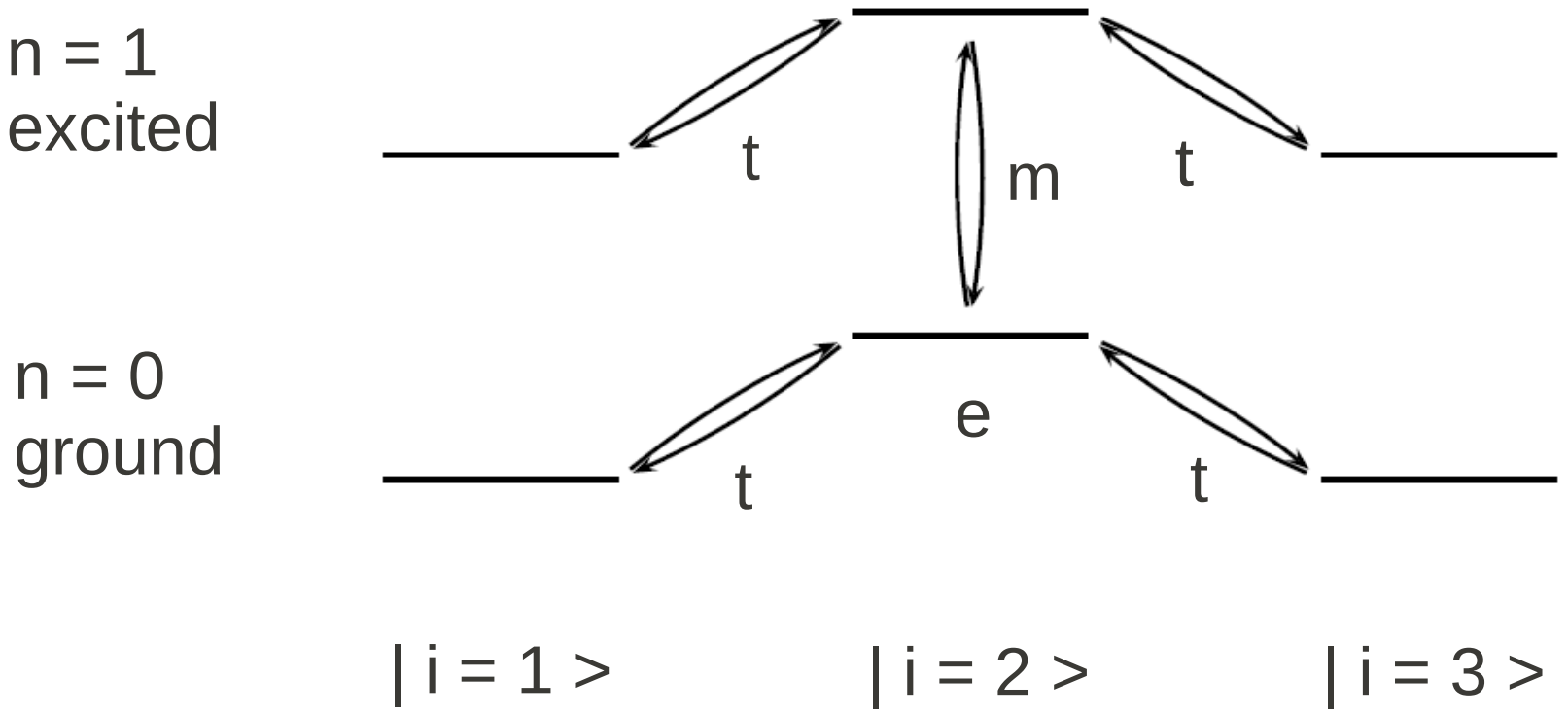
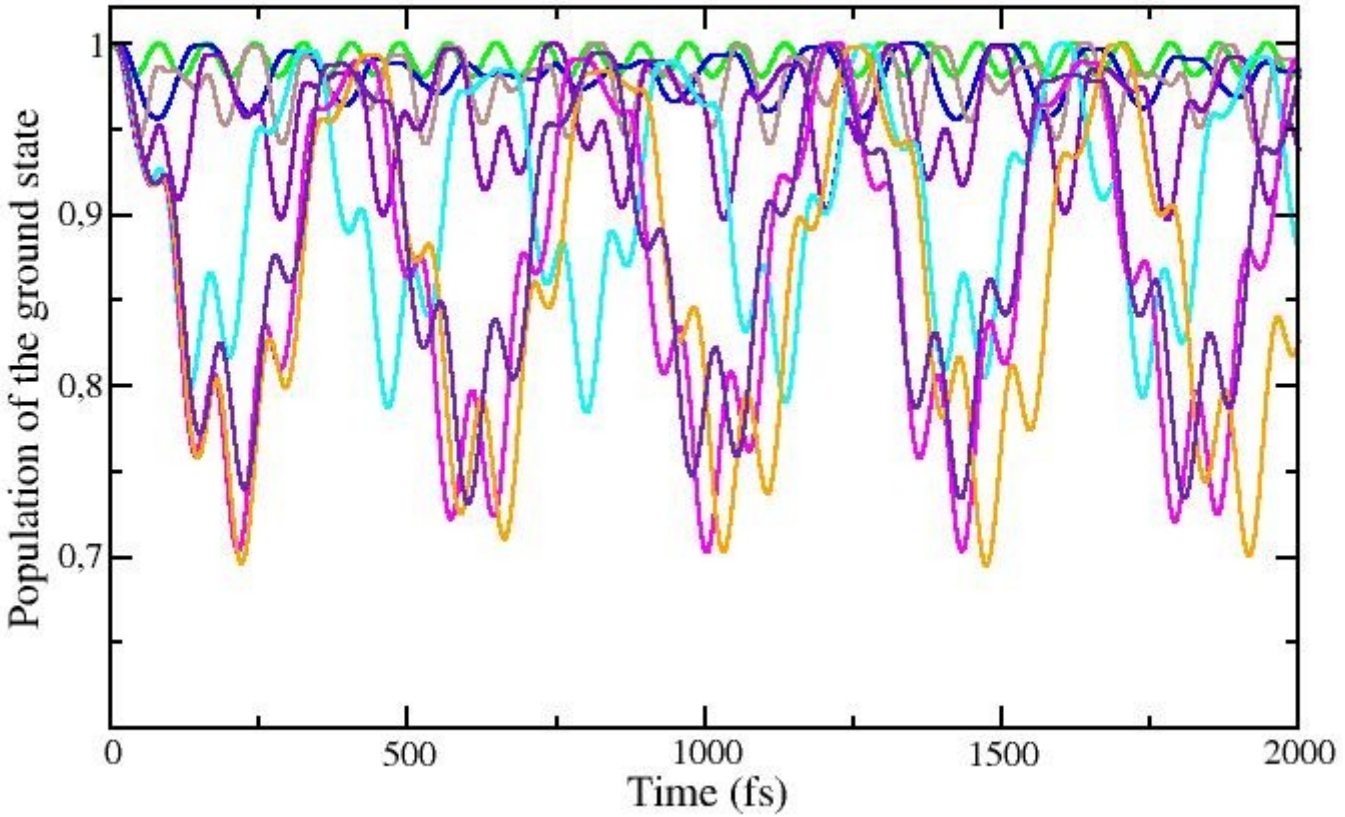
6 states : 3 electronic states coupled to a vibration (n=0, 1)

t : electronic coupling between states in each vibrational subspace
m : vibrational coupling



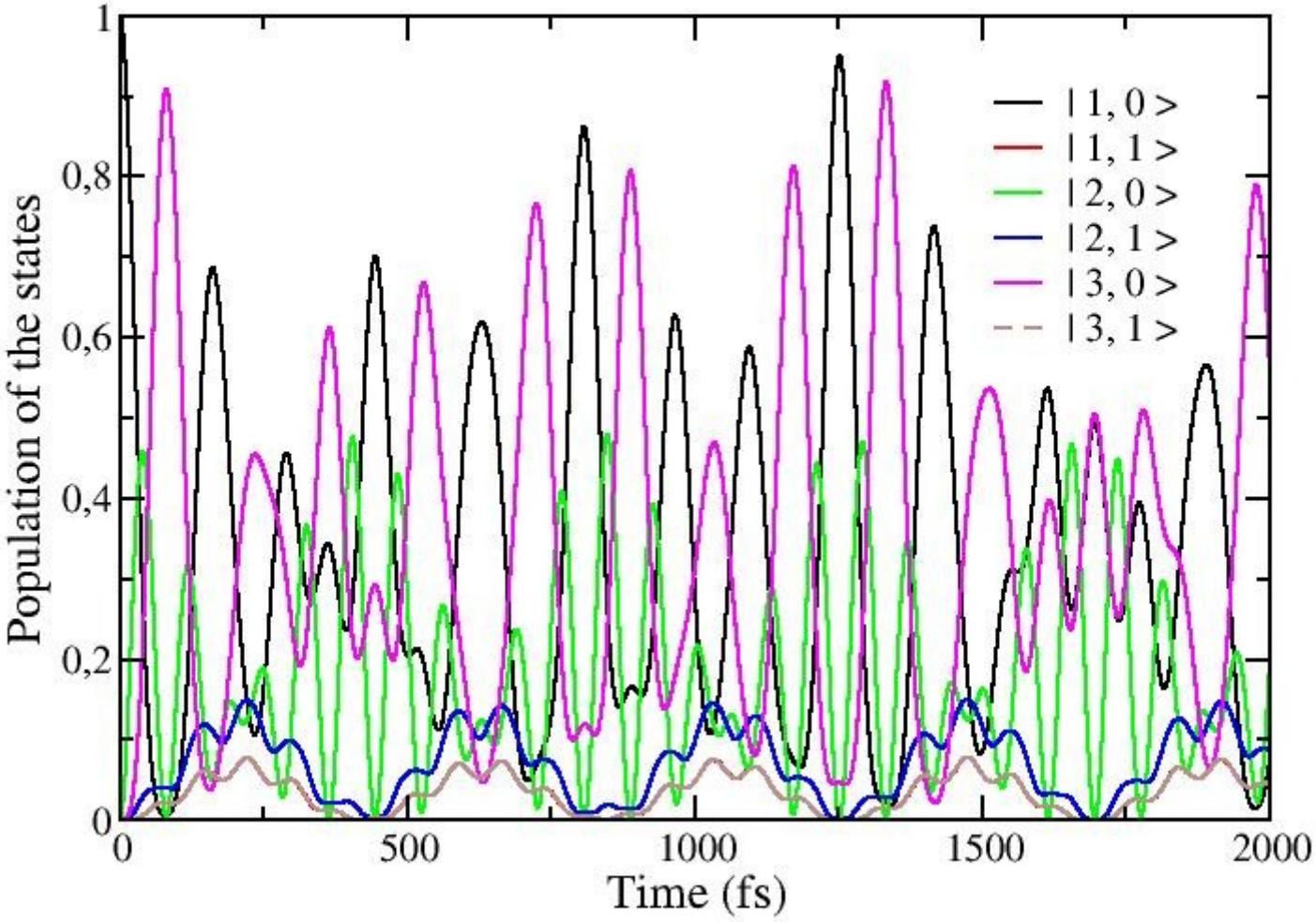
4- A quantum model

Behaviour of the population of the ground state (n=0)



4- A quantum model

Populations of each individual state | i, n >



4- A quantum model

Calculation of the populations

$$\rho_{ii} = \langle i|\hat{\rho}|i\rangle = \langle i||\Psi\rangle\langle\Psi||i\rangle$$

$$\rho_{ii} = \sum_{n,m} C_n(0)C_m^{\star}(0)\langle i||n\rangle\langle m||i\rangle e^{-i(\varepsilon_n-\varepsilon_m)t}$$

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Coefficient associated to frequency ω_{nm}

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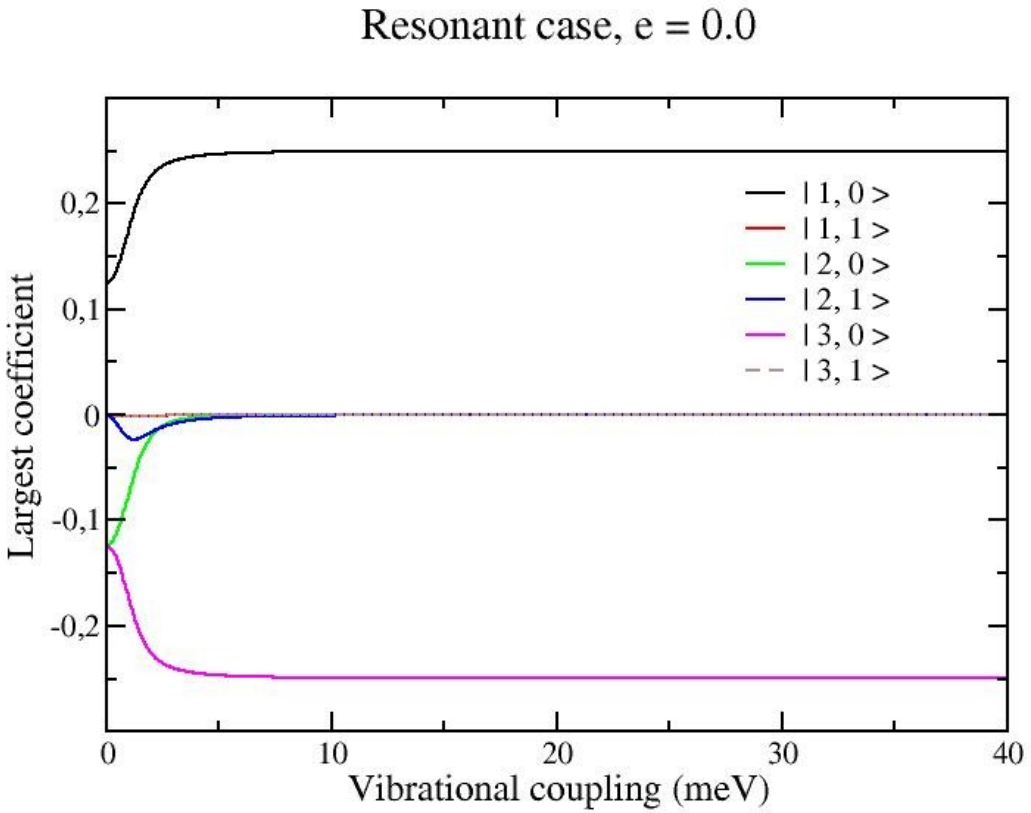
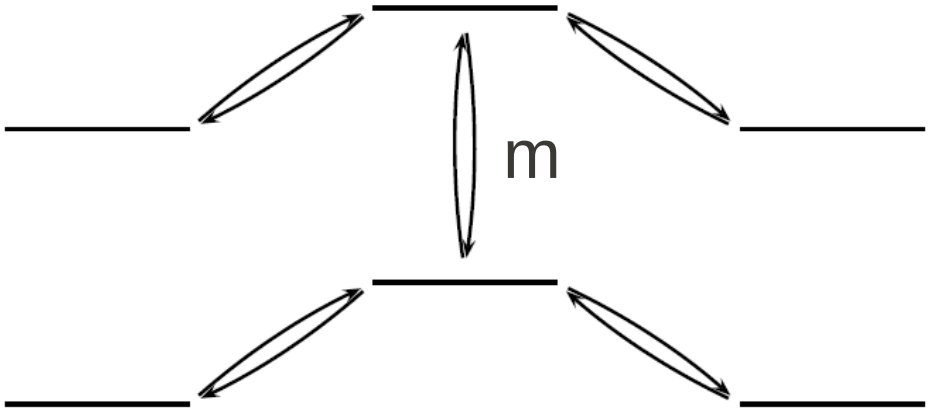
Coefficient associated to frequency ω_{nm}

4- A quantum model

Largest coefficient as a function of the vibrational coupling m





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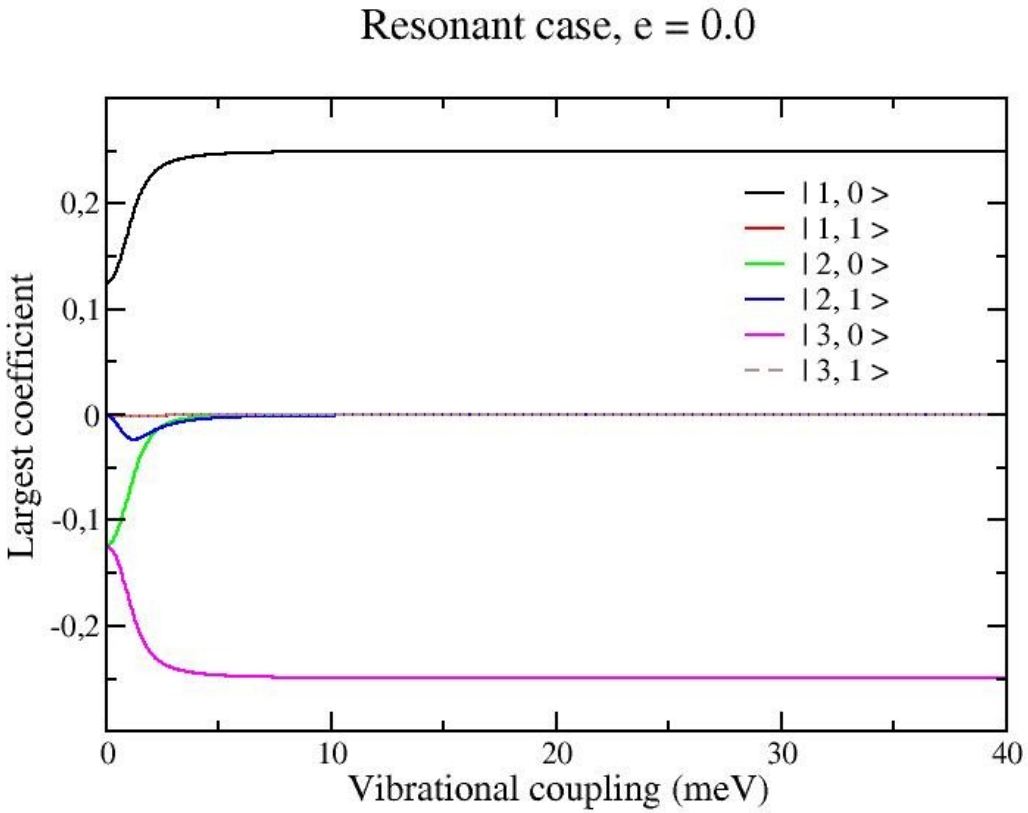

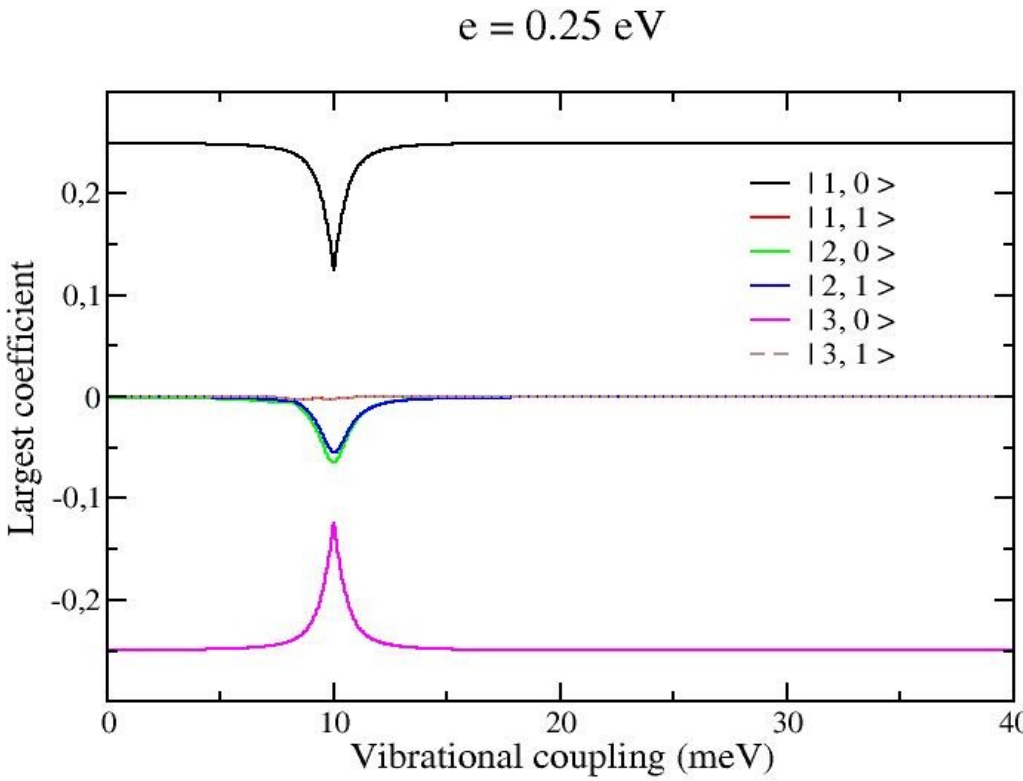

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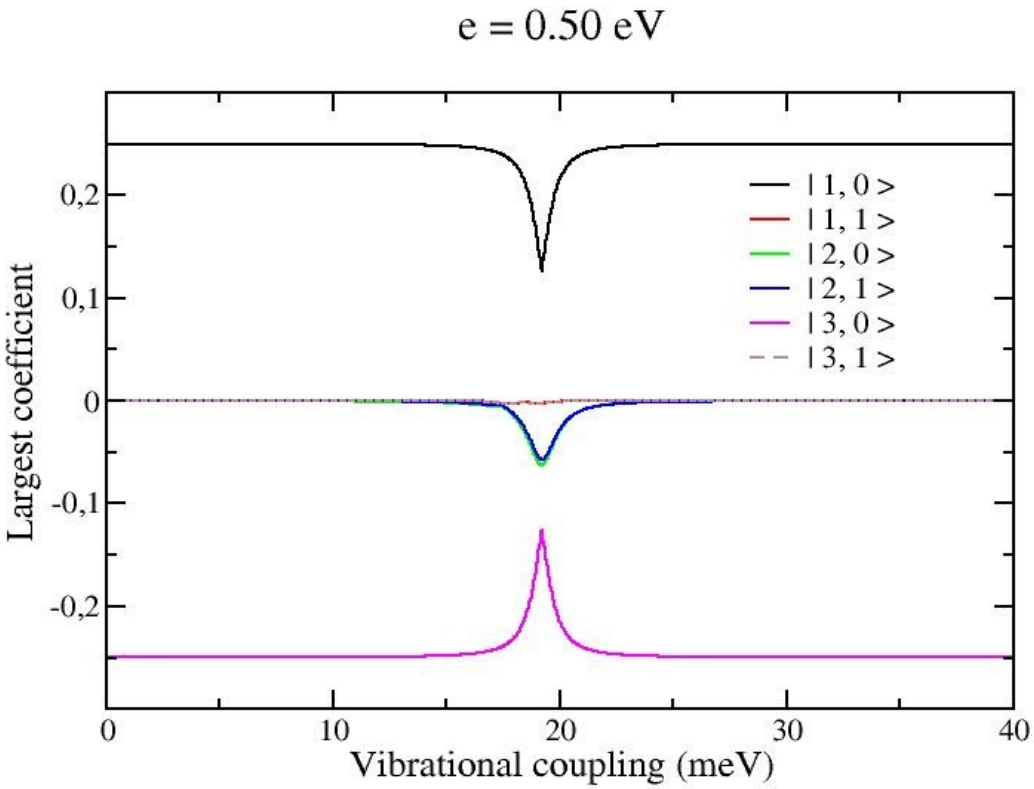
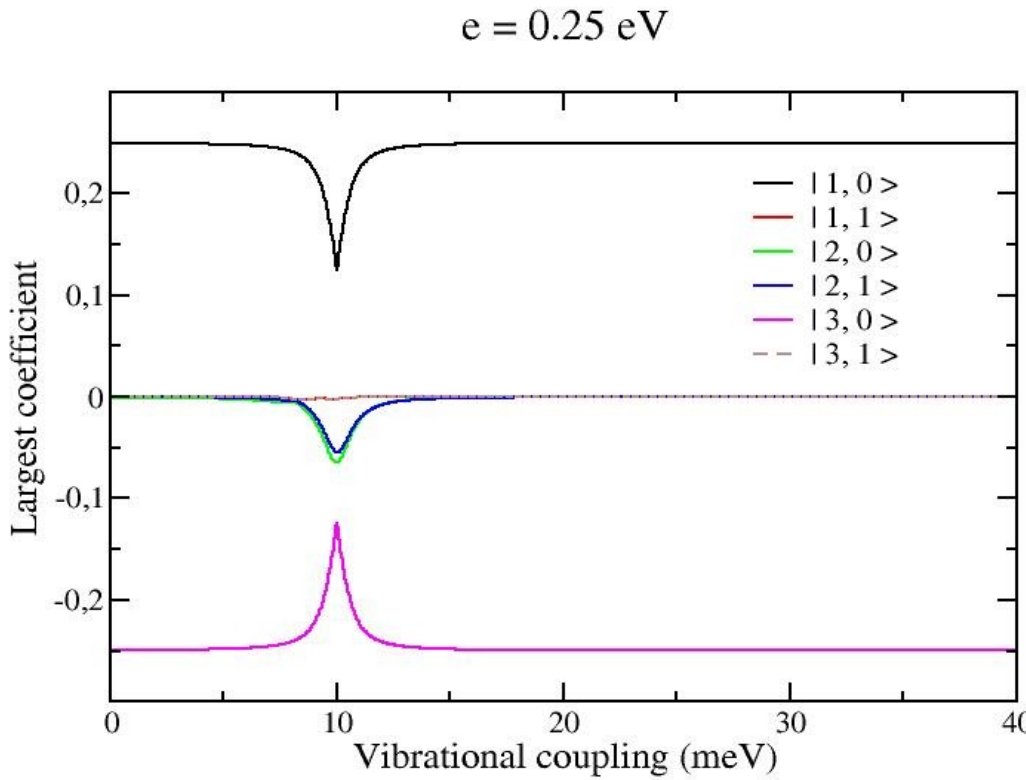
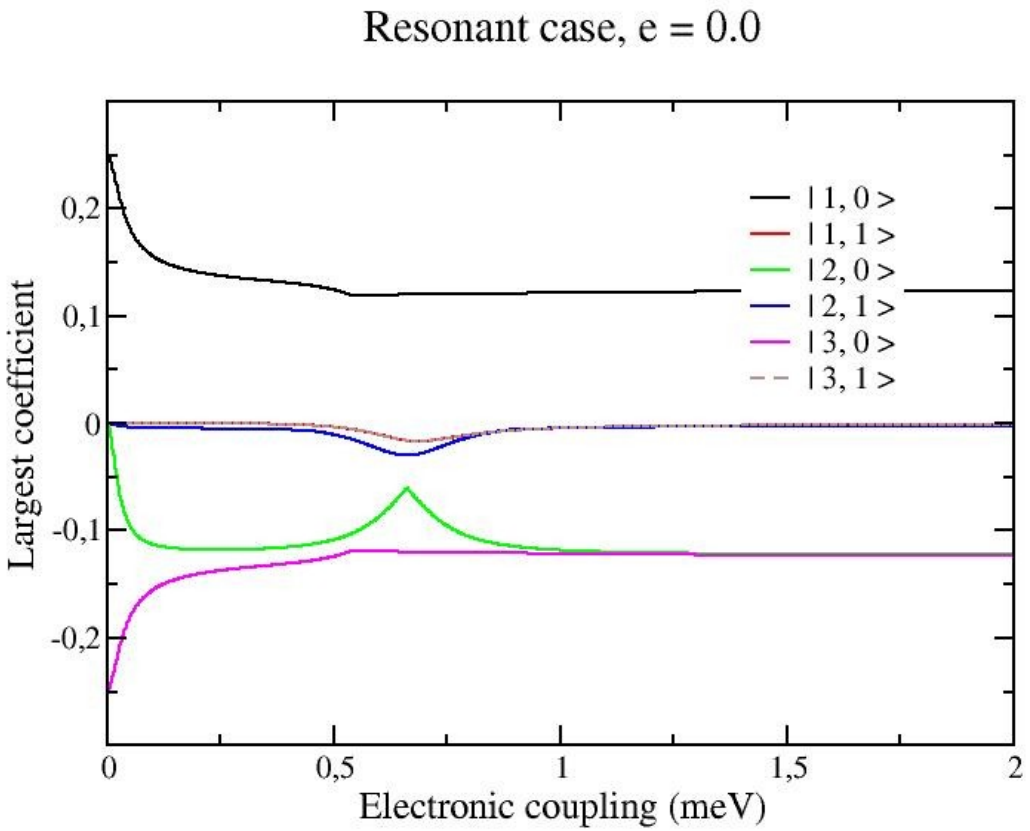
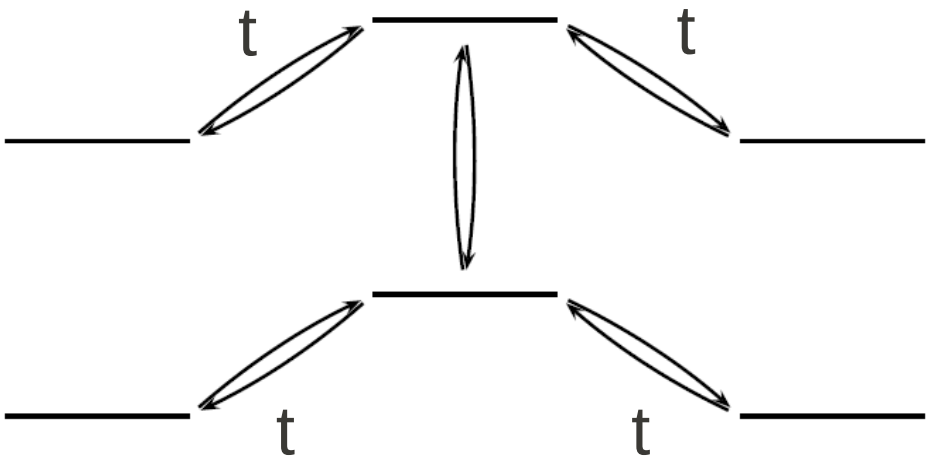




4- A quantum model

Largest coefficient as a function of the electronic coupling t





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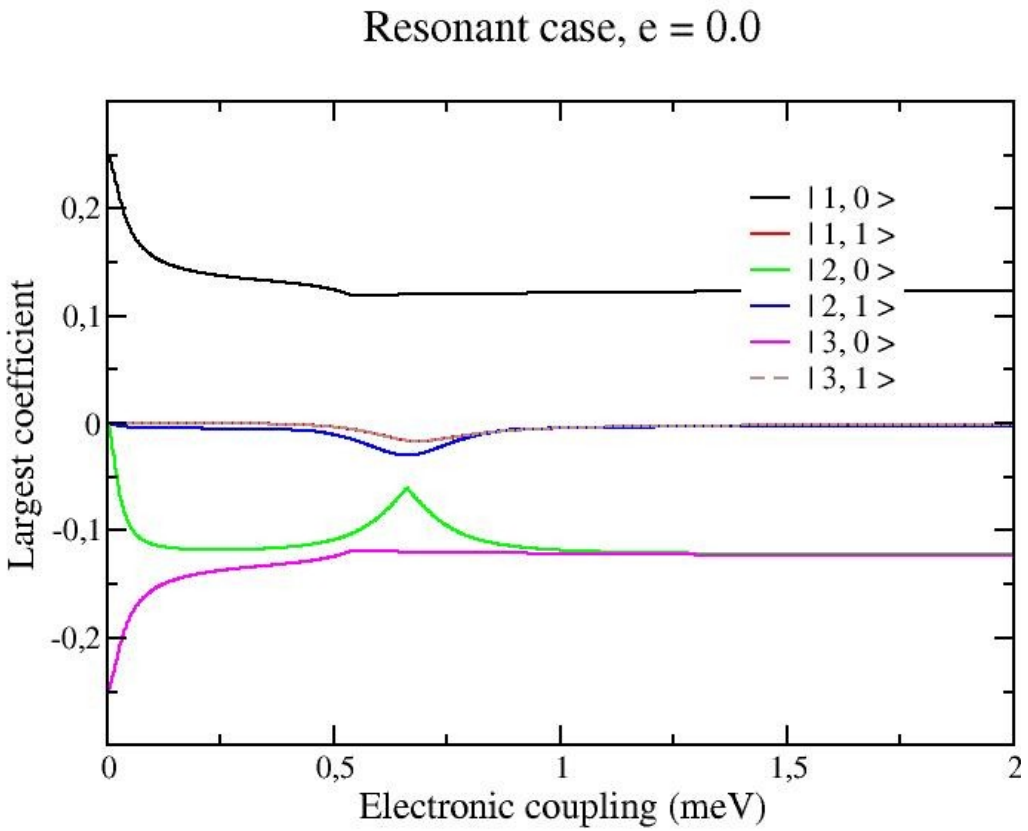

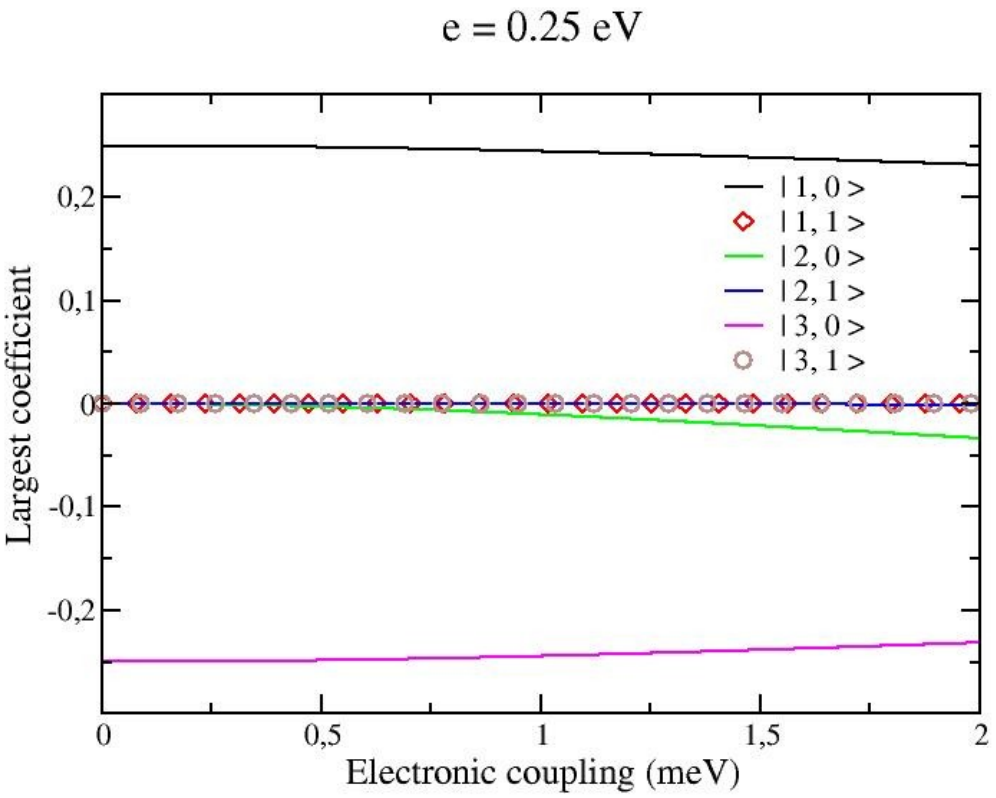

Conclusions

Work in progress

- Find a suitable model to describe dissipation in the quantum/classical model

- Keep exploring the behaviour of the quantum model to find a suitable set of parameters and perhaps a rule

Acknowledgements : Christian Joachim

Funding : AtMol project