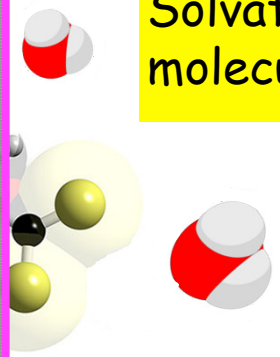
A fluid dynamics approach to the dynamics of microscopic systems

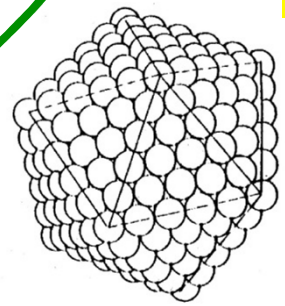
Dynamical description of irradiation and response of
- electrons
- ions
- environment

Irradiation of solvated « bio »molecules
- Microscopic mechanisms
- Role of water environment
- Medical applications
- Society impact



Solvated molecules

Laser irradiations of free clusters
- Huge energy absorption in intense laser fields
- Production of energetic electrons, ions, photons
- Many-body « laboratory »
- Time resolved dynamics



Free clusters

Deposited/embedded species
- Shaping at nanoscale
- Defect formation (electronic devices)
- Chromophore effects and therapy applications

Electron Dynamics

Quantum mean-field (TDLDA)

➢ **Electrons**

- **Big systems : Time Dependent Density Functional Theory (TDDFT)**

Ensemble of orbitals (1 electron) / no correlation $\{\varphi_i(\mathbf{r},t), i=1,\ldots\}$

One body density $\varrho(\mathbf{r},t)=\sum_i |\varphi_i(\mathbf{r},t)|^2$

Effective mean field theory (Kohn-Sham)

$$i\hbar\frac{\partial\varphi_i}{\partial t}=h[\varrho]\varphi_i \qquad h[\varrho]=-\frac{\hbar^2}{2m}\Delta+U_{\rm KS}+U_{\rm ext}(\mathbf{r},t) \qquad U_{\rm KS}=U_{\rm H}+U_{\rm xc}[\varrho]$$

Exch. + Corr.

Kohn-Sham potential | Ions + ext. | Coulomb direct

- **Local Density Approximation (LDA) —> TDLDA**

$i\hbar\partial_t\hat\rho=[\hat h,\hat\rho]$ ($\hat\rho$ 1-body density **matrix**)

➢ **Ions**

- **Explicit classical ions**
- **Detail of structure + ionic Molecular Dynamics (MD)**

Explicit Quantum… but very expansive for large systems

Vlasov and beyond (VUU/BUU)

- At high excitation energy one may use semi classical approximations
  —> Vlasov equation

- Vlasov also provides a sound starting basis for complementing mean-field by dynamical correlations (« Boltzmann-like » collision term)

$$i\hbar\partial_t\hat{\rho} = [\hat{h}, \hat{\rho}]$$ ($\hat{\rho}$ 1-body density **matrix**) TDLDA

$$\hat{\rho}, \hat{h} \xrightarrow{Wigner\ transform} f(\mathbf{r},\mathbf{p},t), h(\mathbf{r},\mathbf{p},t)$$

$$\partial_t f = \{h(\mathbf{r},\mathbf{p},t), f(\mathbf{r},\mathbf{p},t)\}$$ Vlasov

$$\partial_t f = \{h(\mathbf{r},\mathbf{p},t), f(\mathbf{r},\mathbf{p},t)\} + I_{coll}[f]$$ VUU/BUU

- Semi classical kinetic equation (plasmas, nuclear physics…)

But … relatively involved numerics

No big gain in system size nor computation time

From Vlasov to Fluid Dynamics

- Estimate of the local velocity field and stress tensor from Vlasov

$$\mathbf{u}(\mathbf{r}) = \int \frac{\mathbf{p}}{\mathbf{m}} f(\mathbf{r},\mathbf{p},t) d^3r$$

$$\Pi_{ij}(\mathbf{r}) = \frac{1}{m}\int f(\mathbf{r},\mathbf{p},t)\,(\mathbf{p}_i - m\mathbf{u}_i)(\mathbf{p}_j - m\mathbf{u}_j) d^3p$$

Local anisotropy of the velocity field remains small enough

- Fluid dynamics approach (Time Dependent Thomas Fermi)

  Variational principle + wave function ansatz

$$\delta \int dt \langle\psi| i\hbar\partial_t - H|\psi\rangle = 0 \qquad \varphi_i(\mathbf{r},t) = \varphi_i^0(\mathbf{r},t)\exp(i\frac{\hbar}{m}\chi(\mathbf{r},t))$$

$$\mathbf{u}(\mathbf{r},t) = \nabla\chi$$

$$\langle\psi|\hat{T}|\psi\rangle = T^0 + \frac{m\rho u^2}{2}$$

"collective"

"intrinsic"

$$T^0[\varrho] = (3\pi^2)^{2/3}\int \frac{3\hbar^2}{10m}\varrho(\mathbf{r},t)^{5/3} d^3r$$

Thomas Fermi approximation (local Fermi gas)

From Vlasov to Fluid Dynamics (cont'd)

- Fluid dynamics equations (Time Dependent Thomas Fermi)

$$\frac{\partial \varrho}{\partial t} + \nabla.(\varrho \mathbf{u}) = 0$$

$$m\frac{\partial \chi}{\partial t} + \frac{m}{2}(\nabla\chi)^2 + \frac{\delta U}{\delta \varrho} + \frac{\delta T}{\delta \varrho} = 0$$

- Resolution via Madelung transform (ONE wavefunction)

$$\Phi(\mathbf{r},t) = \sqrt{\varrho(\mathbf{r},t)}\exp(i\frac{\hbar}{m}\chi(\mathbf{r},t))$$

$$i\hbar\frac{\partial \Phi}{\partial t} = -\frac{\hbar^2}{2m}\Delta\Phi + (\frac{\delta U}{\delta \varrho} + \frac{\delta T^0}{\delta \varrho})\Phi + \frac{\hbar^2}{2m}\frac{\Delta\sqrt{\varrho}}{\sqrt{\varrho}}$$

Full 3D time dependent quantum package
but for ONLY 1 wave function —> cheaper

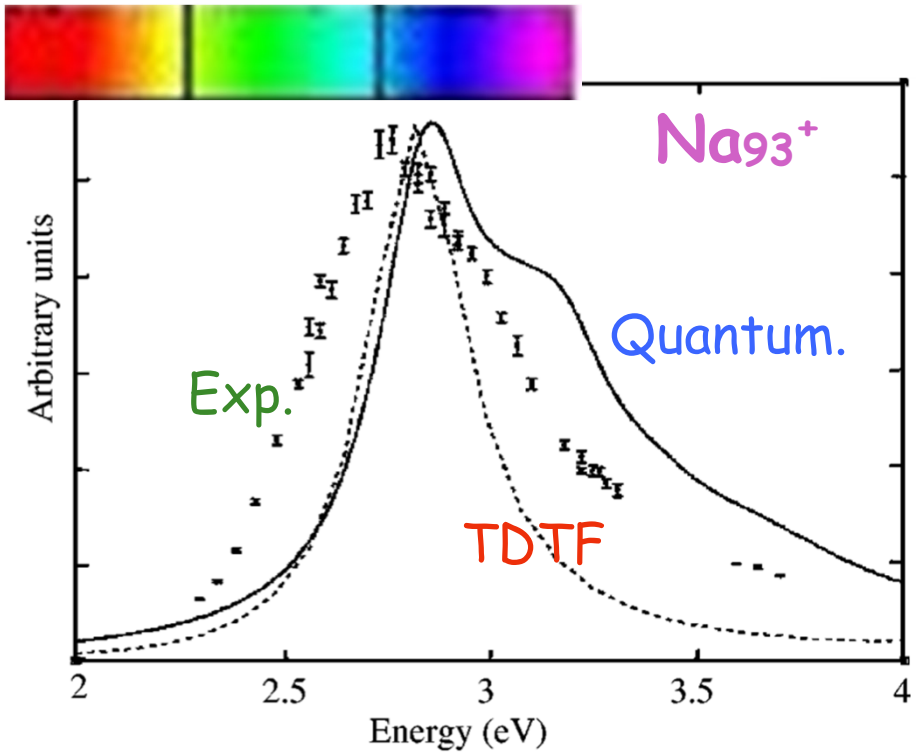
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Check of validity : optical response

- Optical response (collective oscillations of electrons with respect to ionic background)
—> low energy

- Basically classical effect at least for gross features

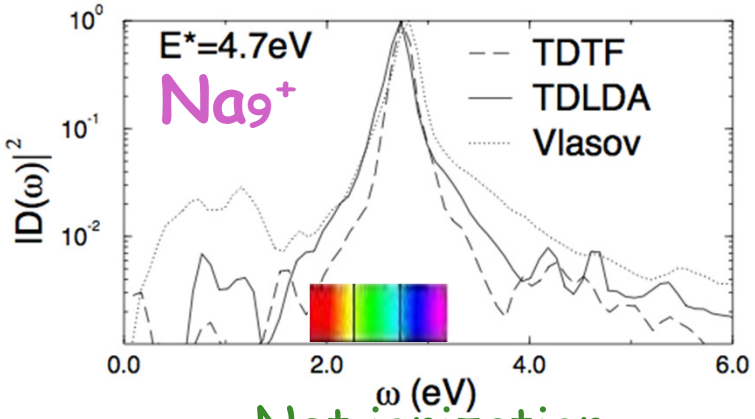
- Entrance channel for coupling to light in many dynamical regimes



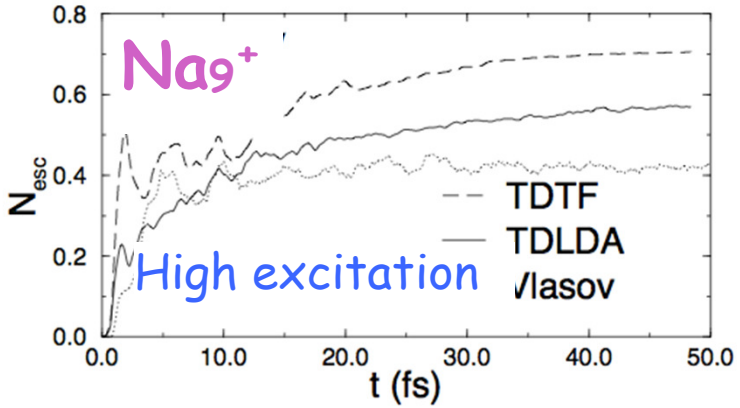
↳ Good (sufficient) agreement

Behavior in the non linear domain

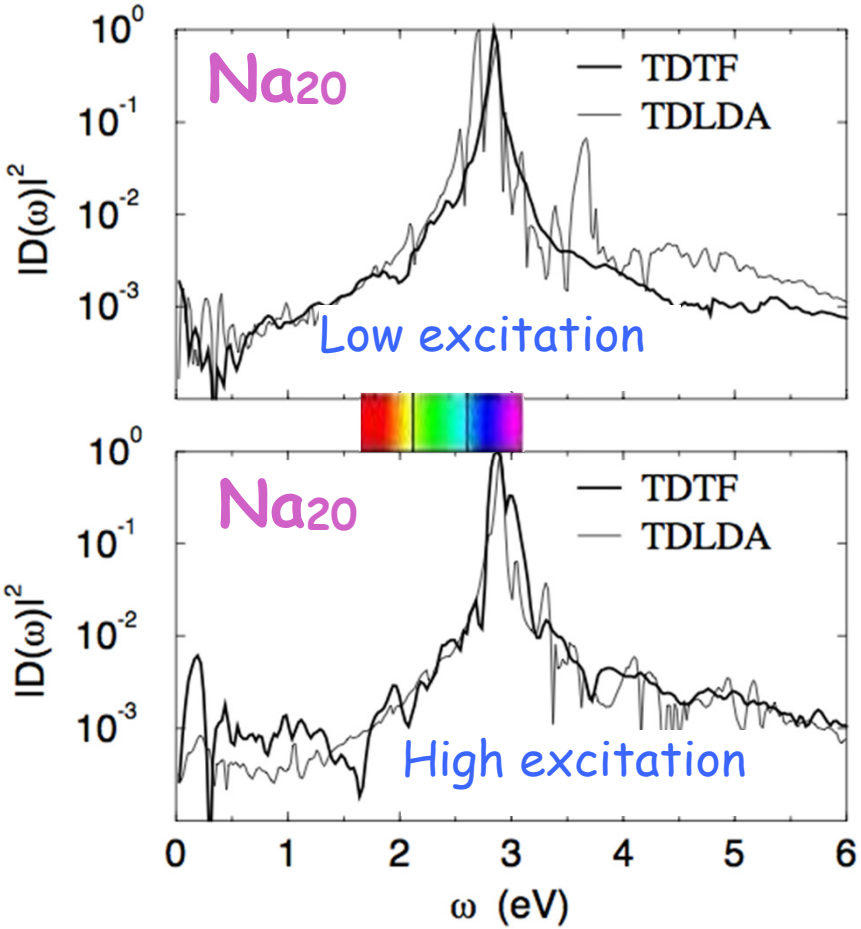
Optical response



Net ionization



Optical response



Even better at high excitation energy…

Conclusions and perspectives

- Fluid dynamics (Time Dependent Thomas Fermi) provides a sound description of electron dynamics in simple clusters for various dynamical regimes
- Possible applications to irradiation dynamics of nano objects
- Numerically cheap solution for large (nano) systems
- Quantum softwares may provide a robust tool for solving fluid dynamics equations
- Dissipative extensions to be explored


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for
your
attention

Thank you too…

People

P. G Reinhard
A. Domps

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