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Comparison of time steppers for Laser Ablation

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Outline

- About Laser Ablation (LA)
- Phenomena involved in LA
- Hydrodynamic model
- Collisional Radiative model for plasma formation
- Simulations: effect of intensity and of wave length
- Simulations: timings of 17 ODE solvers
- Conclusions

Laser Ablation

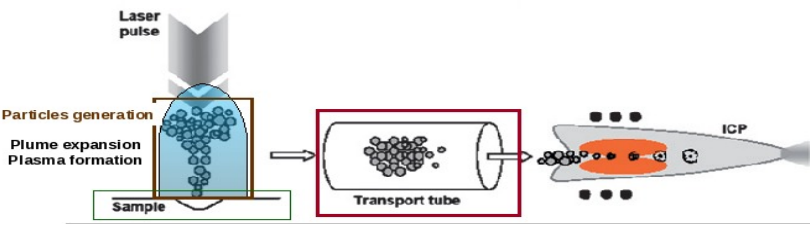
Laser Ablation is

Process of removing material using a pulsed laser beam

Many application areas, among which are:

- cutting, machining, drilling, engraving
- bonding, coating, surface cleaning
- surgery, dentistry, ... lasik, hair/tatoo removal
- Laser-induced breakdown spectroscopy
- LA - Inductively coupled plasma mass spectrometry ...

Laser Ablation - Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS)



to detect ultra-trace elements, as low as 2 parts per 100 billion!
Widely used in chemistry, medicine, archaeology, geology, materials science, ...

Processes in Laser Ablation

What happens during Laser Ablation:

- solid target heats up, a thin layer melts
- melt vaporizes at high T, creating a plume of vapor
- which rapidly expands (and cools somewhat)
- electrons/ions dissociate creating plasma

What happens where:

- **in target:** heat conduction / melting / vaporization
  coupled to electric field (thermionic emission)
- **over target:** Knudsen transition layer
- **in plume:** plasma formation, expansion, laser absorption,
  shielding of target, recondensation
- Near T_{crit}: phase explosion (vapor and liquid dropplets)

Desirable features to be modeled:

- seamless treatment of heating / melting / vaporization
- T-dependence of thermophysical properties,
  and of reflection & absoption coeffs
- vapor as non-ideal gas, need EoS up to critical T ($\sim$ 8000 K)
- spatially non-uniform laser pulse
- target - plume coupling via Knudsen Layer for
  subsonic/sonic vaporization, condensation
- determine T_{vap} , surface recession velocity v_{rec}
- plume expansion/recondensation, in vacuum or in background gas
- plasma formation (ions and electrons)
- absorption of laser energy in plume / shielding of target
- capture (very) strong shocks via high resolution schemes
- in 2D (axisymmetric) and 3D (but first in 1D !!!)
- thermionic emission, Coulomb explosion: coupling to electric field

Additional Challenges

Additional challenges in modeling Laser Ablation:

- extreme space and time scales
- extreme gradients: T may rise to thousands of degrees locally
- extreme variation in thermo-physical properties :
  e.g. $k(T)$ may vary by 12 orders of magnitude
- coupling vaporization to gas flow via Knudsen Layer
- track many species of ions, electrons in plasma
- account for phase explosion (spinodal decomposition)
- need extensive thermo-physical data :
  $\rho(T, P)$, $c_p(T)$, $k(T)$,
  phase diagram for **solid, liquid, vapor**
  over the range 300 K to critical T ($\sim$ 8000 K)

1D model for target

Target heating / melting / vaporization
described by Heat Conduction equation for *internal energy U*
in frame attached to receding surface moving with velocity v_{rec},
valid throughout the target, irrespectively of phase.

$$\frac{\partial U(z,t)}{\partial t} - v_{rec}(t)\frac{\partial U(z,t)}{\partial z} = \frac{\partial}{\partial z} k(T) \frac{\partial T(z,t)}{\partial z} + S_{las}(z,t),$$

with laser source

$$S_{las}(z,t) = I_0 \cdot (1 - R(T))\,\alpha(T)\,I(t)\,\exp(-\alpha(T)z).$$

$R(T)$ = reflectivity , $\alpha(T)$ = absorption coefficient
from Drude model via Wiedemann-Franz law,
$I(t)$ = laser intensity, Gaussian in time.

$U(T, P, \rho)$ updated by PDE, then *phase*, ρ, T
found from **Equation of State** (ITTEOS code).

Thin transition layer (few mean free paths) between liquid and vapor where molecules are not in translational equilibrium.

T, P, ρ, v undergo jumps governed by local Mach number M.
We use Knight's relations for vaporization into a background gas:

$$\frac{T_K}{T_s} = \left[\sqrt{1 + \pi \left(\frac{m}{2} \left(\frac{\gamma - 1}{\gamma + 1} \right) \right)^2} - \sqrt{\pi} \frac{m}{2} \left(\frac{\gamma - 1}{\gamma + 1} \right) \right]^2,$$

$$\frac{P_K}{P_s} = \sqrt{\frac{T_K}{T_s}} \left[\left(m^2 + \frac{1}{2} \right) e^{m^2} \mathrm{erfc}(m) - \frac{m}{\sqrt{\pi}} \right] + \frac{1}{2} \left[1 - \sqrt{\pi} e^{m^2} \mathrm{erfc}(m) \right],$$

$$m = \frac{v_K}{\sqrt{2 R T_K}}, \quad M = \frac{v_K}{\sqrt{\gamma R T_K}}.$$

$P_K/P_s \leq 1$ for vaporization ($M \geq 0$), else
$P_K/P_s = 0.95 e^{2.42|M|} > 1$ for condensation ($M < 0$).

Knudsen Layer 2

From energy and mass conservation and the KL jump conditions, we determine the following at the "outside"(plume side) of the KL:

- Mach number (subsonic/sonic vaporization or condensation)
- recession velocity of melt surface
- vaporization T
- saturation Pressure
- vapor density
- vapor velocity ($\rightarrow$ inflow BC for CFD in plume)

CFD in plume

Conservation of mass, momentum, energy, species (vapor, background gas). So we solve **compressible Navier–Stokes with species and total energy**, coupled to **EoS** $U(T, P, \rho)$ and **phase diagram**, up to T_{crit}.

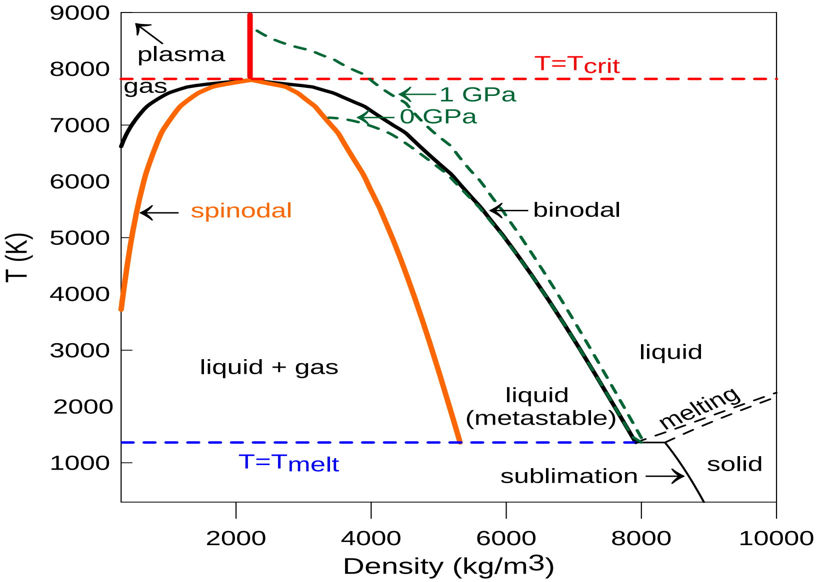
$$\frac{\partial \rho}{\partial t} = -\frac{\partial \rho v}{\partial z}$$

$$\frac{\partial \rho_{vap}}{\partial t} = -\frac{\partial \rho_{vap} v}{\partial z} - \frac{\partial}{\partial z} j_m^x$$

$$\frac{\partial \rho v}{\partial t} = -\frac{\partial}{\partial z}\left(\rho v^2 + P + \frac{\partial}{\partial z}\tau_{zz}\right)$$

$$\frac{\partial \rho \varepsilon}{\partial t} = -\frac{\partial}{\partial z}\left[\rho \varepsilon v + P v\right] - \frac{\partial}{\partial z}\left(q + \tau_{zz}\right) + S_{\mathrm{IB}} + S_{\mathrm{MPI}} - S_{rad}$$

These are coupled to the (outer side of) Knudsen Layer...



9000
8000
7000
6000
5000
4000
3000
2000
1000
T (K)
plasma
gas
T=Tcrit
1 GPa
0 GPa
spinodal
binodal
liquid + gas
liquid
liquid
(metastable)
melting
T=Tmelt
sublimation
solid
2000
4000
6000
8000
10000
Density (kg/m3)

explosive boiling near T_{crit}

When temperatures around $0.9T_{crit}$ are reached:

- large fluctuations in density and entropy occur
- hot liquid metal enters metastable state
  (btn the binodal and the spinodal of phase diagram)
- spinodal decomposition drives system back into equilibrium state
- fast increase of homogenous nucleation rate and bubble growth results in ejection of a mixture of vapor and liquid droplets.
- Homogenous nucleation rate J_c from Volmer-Döring theory
- critical radius for nucleation: $R_{cr} = 2\sigma(T)/(P_s(T) - P_{amb})$
- surface tension obtained from extended Eötvös rule

Species in plasma

Considered as binary mixture of vapor and background gas consisting of ions, electrons, and population of excited levels.

- when $T_{surf} > T_{boil}$: optical breakdown (plasma forms) via Collisional and Radiative (C-R) processes.
- soon after, Local Thermodynamic Equilibrium attained: ODEs turned off and LTE modelled by Saha-Eggert equations.
- Saha eqs and corresponding closure relations solved iteratively by a Newton-Raphson method to calculate molar fractions of ions at each T.
- Spectroscopic data from NIST database.

Collisional-Radiative model is a system of ODEs describing:

- single- and multi-photon ionization,
- radiative decay electron impact excitation and ionization,
- respective recombination reactions, Inverse Bremsstrahlung.

I. Main radiative processes

- Laser-induced photo ionization:

$$Cu_i{}^k + m\hbar\omega_{\mathrm{laser}} \rightarrow Cu_j{}^{k+1} + e$$

$$\frac{dN_j^k}{dt} = \sum_{i=1}^{n(k-1)} k_{PI,i,j}^{k-1} N_i^{k-1} - \sum_{i=1}^{n(k+1)} k_{PI,i,j}^{k+1} N_j^k$$

$$\begin{aligned} S_{PI,j}^k &= \sum_{i=1}^{n(k-1)} k_{PI,i,j}^{k-1} N_i^{k-1} (m\hbar\omega_{\mathrm{laser}} - \Delta E_{ij}) \\ &+ \sum_{i=1}^{n(k+1)} k_{PI,i,j}^{k+1} N_j^k (m\hbar\omega_{\mathrm{laser}} - \Delta E_{ij}) \end{aligned}$$

Collisional Radiative model (cont'd)

- Spontaneous decay:

$$Cu_j{}^k \to Cu_i{}^k + \hbar\omega_{ij}$$

$$\frac{dN_j^k}{dt} = \sum_{i=j+1}^{n(k)} k_{dec,i,j}^k N_i^k - \sum_{i=1}^{j-1} k_{dec,i,j}^k N_j^k$$

II. Main collisional processes

- Collisional excitation /de-excitation:

$$Cu_i{}^k + e \leftrightarrow Cu_j{}^k + e$$

$$\frac{dN_j^k}{dt} = \sum_{i=1}^{j-1} \left(k_{exc,i,j}^k N_i^k - k_{dexc,i,j}^k N_j^k \right) N_e + \sum_{i=j+1}^{n(k)} \left(k_{dexc,i,j}^k N_i^k - k_{exc,i,j}^k N_j^k \right) N_e$$

$$S_{ex,j}^k = -\sum_{i=1}^{j-1} \left(k_{exc,i,j}^k N_i^k - k_{dexc,i,j}^k N_j^k \right) \Delta E_{ij} N_e + \sum_{i=j+1}^{n(k)} \left(k_{dexc,i,j}^k N_i^k - k_{exc,i,j}^k N_j^k \right) \Delta E_{ij} N_e$$

- Electron impact ionization/ three body recombination:

$$Cu_i{}^k + e \leftrightarrow Cu_1{}^{k+1} + e$$

$$\begin{aligned}
\frac{dN_j^k}{dt} = & \sum_{i=1}^{n(k-1)} \left(k_{ei,i,j}^{k-1} N_i^{k-1} - k_{tbod,i,j}^{k-1} N_j^k N_e \right) N_e \\
& + \sum_{i=1}^{n(k+1)} \left(k_{tbod,i,j}^{k+1} N_e N_i^{k+1} - k_{ei,i,j}^{k+1} N_j^k \right) N_e \\
S_{ei,j}^k = & - \sum_{i=1}^{n(k-1)} \left(k_{ei,i,j}^{k-1} N_i^{k-1} - k_{tbod,i,j}^{k-1} N_j^k N_e \right) \Delta E_{ij} N_e \\
& + \sum_{i=1}^{n(k+1)} \left(k_{tbod,i,j}^{k+1} N_e N_i^{k+1} - k_{ei,i,j}^{k+1} N_j^k \right) \Delta E_{ij} N_e
\end{aligned}$$

III. The ODEs System:

$$
\begin{aligned}
\frac{dN_j^k}{dt} \quad &= \sum_{i=1}^{j-1} \left(k_{exc,i,j}^k N_i^k - k_{dexc,i,j}^k N_j^k \right) N_e \\
&+ \sum_{i=j+1}^{n(k)} \left(k_{dexc,i,j}^k N_i^k - k_{exc,i,j}^k N_j^k \right) N_e \\
&- \sum_{i=1}^{j-1} k_{dec,i,j}^k N_j^k + \sum_{i=j+1}^{n(k)} k_{dec,i,j}^k N_i^k \\
&+ \sum_{i=1}^{n(k-1)} \left(k_{ei,i,j}^{k-1} N_i^{k-1} - k_{tbod,i,j}^{k-1} N_j^k N_e \right) N_e \\
&+ \sum_{i=1}^{n(k+1)} \left(k_{tbod,i,j}^{k+1} N_e N_i^{k+1} - k_{ei,i,j}^{k+1} N_j^k \right) N_e \\
&+ \sum_{i=1}^{n(k-1)} k_{PI,i,j}^{k-1} N_i^{k-1} - \sum_{i=1}^{n(k+1)} k_{PI,i,j}^{k+1} N_j^k
\end{aligned}
$$

ODE system (cont'd)

$$N^k = \sum_{j=1}^{n(k)} N_j^k, \qquad N_h = \sum_{k=0}^{2} N^k, \qquad N_e = \sum_{k=0}^{2} k N^k$$

$$\begin{aligned} \frac{d}{dt}\left(\tfrac{3}{2} k T_e N_e\right) &= -3\tfrac{m_e}{m_h}\left(T_e - T_h\right)\left(\nu_{en} + \nu_{ei}\right) N_e \\ &+ S_{IB} + \sum_{k=0}^{2} \sum_{j=1}^{n(k)} \left(S_{ei,j}^k + S_{ex,j}^k + S_{PI,j}^k\right) \end{aligned}$$

$$\frac{d}{dt}\left(\tfrac{3}{2} k T_h N_h\right) = 3\frac{m_e}{m_h}\left(T_e - T_h\right)\left(\nu_{en} + \nu_{ei}\right) N_e$$

ODE system (cont'd)

Concise (computational) representation of the ODE system

- Collect the n unknowns into a vector $y \in \mathbb{R}^n$,
- and the coefficients/parameters into a vector $p \in \mathbb{R}^m$. They depend strongly on temperature and reflection coefficients.

Then the collisional radiative model constitutes a system of n first order ODEs for y, containing m parameters p, of the form:

$$\frac{dy}{dt} = f(t, y(t,p), p),\ 0 < t \leq t_{max},\ y \in \mathbb{R}^n,\ p \in \mathbb{R}^m$$

- This system can be solved numerically by any ODE integrator, and we present several.
- Evaluation of $f(t, y(t,p), p)$ is very expensive ...
- In our simulations, $n = 31$ and $m = 4$.

Computational Challenges

- highly nonlinear, strongly coupled system
- multi-phase problem
- moving boundaries (phase fronts)
- extreme space and time scales
- extreme temperature, pressure, density gradients
- hydrodynamic shocks
- ODE system requires smaller Δt than the PDEs . **Is it stiff ?**

Computational Approach

Target heating:

- update U from conservation law
- determine phase fractions and T from EoS

Vapor expansion: if $T \geq T_{boil}$, compute:

- T_{vap}, P_{sat}, v_{rec} using KL jump conditions

Plasma formation: If $T_{melt} \leq T < T_{crit}$

- solve ODEs (breakdown process) and compute laser absorption coefficients

ODE system contains (various) fast scales and requires smaller time-step than the PDEs. We use $\Delta t_{ODE} = \Delta t_{CFL}/factor$, and report the *factor* used by each solver.

Computational Approach (cont'd)

CFD in plume:

- from energy and mass conservation and KL jump conditions, determine M, v_{rec}, T_{vap}, P_{sat}, ρ_{vap}, v_{vap} outside of KL $\Longrightarrow$ inflow BC for Navier-Stokes equations in plume.
- solve the NS with species and energy.
- estimate the growing plume height by integrating speed of sound $h = \int a(t)dt$, and extend computational domain for NS equations.
- update iteratively

Numerical Schemes

Spatial Discretization

- Finite Volume discretization in space (2nd order)
- Non-uniform grid in target, denser near the surface
- Expanding grid (adaptively) in plume region

Time Steppers

- Explicit Euler in time for PDEs
- various ODE solvers for ODEs: **will compare 17 schemes**
  - Explicit Euler
  - Explicit RK2, RK3, RK4, RKFB4, DoPri5
  - Explicit PEER (adaptive) order 7,8,9
  - Intel DODE (adaptive, high order): dodesol, rkm9st, mk52lfn
  - VODE (variable order,adaptive) Adams:10,12, BDF:20,22

The numerical code is implemented in Fortran 90

Ablation of Cu in He background gas
initially at T=300K, P=1atm.

- target thickness: $12\mu m$, nonuniform grid of 2000 CVs along direction of laser beam, denser near the surface
- plume regions: $\Delta z = 500$nm, adaptively increase with growing plum height
- simulation time: $t_{max} = 50$ns, $\Delta t_{CFL} = 10^{-13}$s
  Δt is divided by a *factor* of up to 500 for ODEs.
- Gaussian (in time) laser source (of FWHM=6 ns)

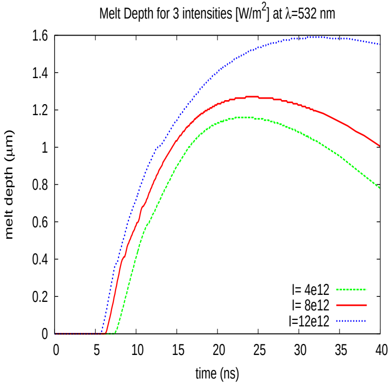
$$S(t,z) = (1-R)\,\alpha\,\beta\,I_0\,e^{-\alpha z-(t_{max}-t)^2/2\sigma^2}$$

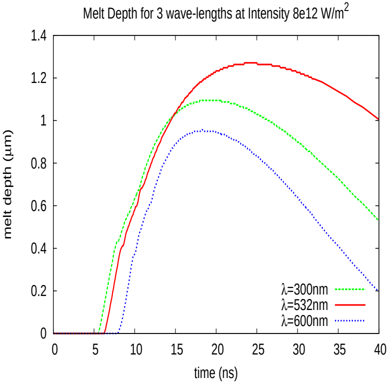
$R(T)$, $\alpha(T)$, $\beta(T)$: reflection, absorption, shielding coefficients of the target.

Simulations shown

- for 3 intensities: $I_0 =$ 4.e12, 8.e12, 12.e12 W/m^2
- and 3 wave-lengths: $\lambda =$ 300 532 600 nm.

Effect of *Intensity* and *wavelength* on: **melt-depth**

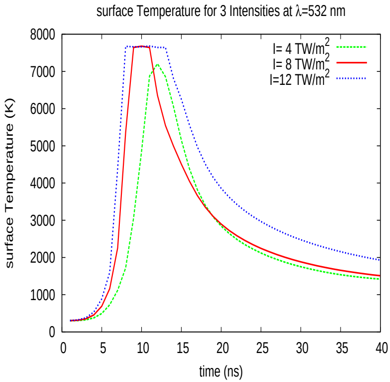


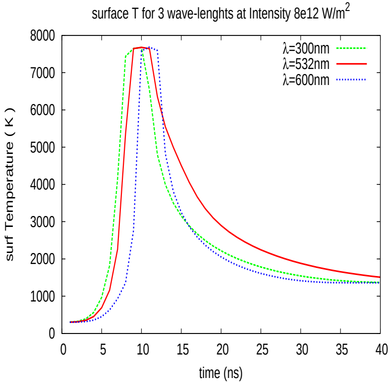


Dependence on I_0 for $\lambda = 532$nm

Dependence on λ for $I_0 = 8.e12\, W/m^2$

Effect of *Intensity* and *wavelength* on: **surface Temperature**

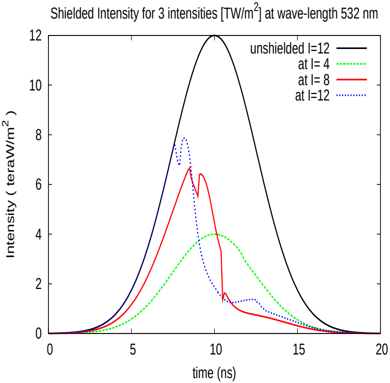


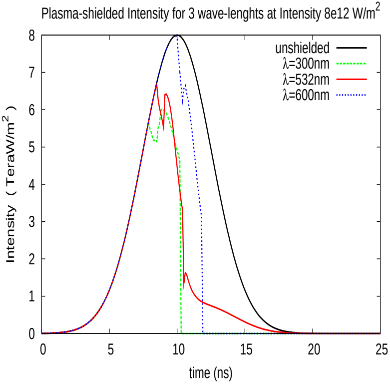


Dependence on I_0 for $\lambda = 532$nm

Dependence on λ
for $I_0 = 8.e12\ W/m^2$

Effect of *Intensity* and *wavelength* on: **plasma shielding of target**



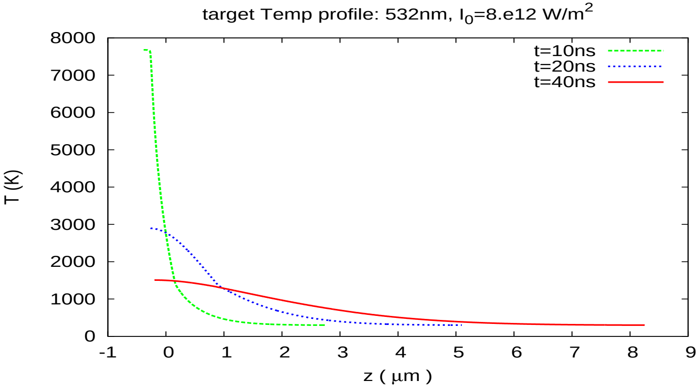


Dependence on I_0 for $\lambda = 532$nm

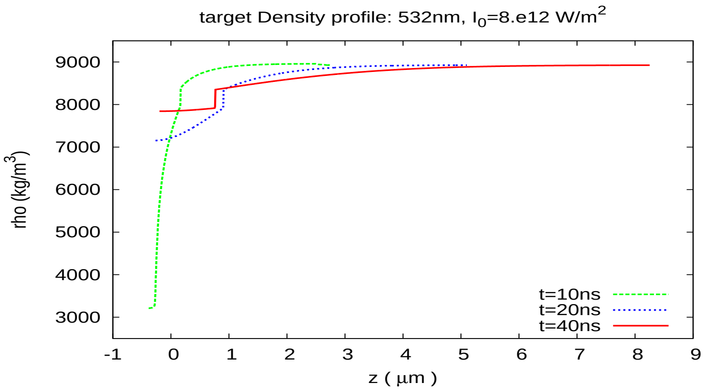
Dependence on λ for $I_0 = 8.e12\ W/m^2$

Profiles

Temperature profiles in target at 3 times
for λ=532nm, I_0=8.e12 W/m^2



Density profiles in target at 3 times
for λ=532nm, I_0=8.e12 W/m^2



Comparison of 17 ODE time-steppers

- Explicit Euler
- Explicit RK2, RK3, RK4
- Explicit, adaptive RKFB4, DoPri5 (Dormand-Prince)
- Explicit, adaptive PEER of orders 7, 8, 9
- Intel DODE (adaptive, high order): dodesol, rkm9st, mk52lfn:
  dodesol: all-purpose, stiff, non-stiff, stability control
  rkm9st: explicit, Merson 4th, multistage $\leq$ 9 stages
  mk52lfn: implicit, L-stable(5,2), for stiff
- VODE (variable order, adaptive) Adams:10,12, BDF:20,22

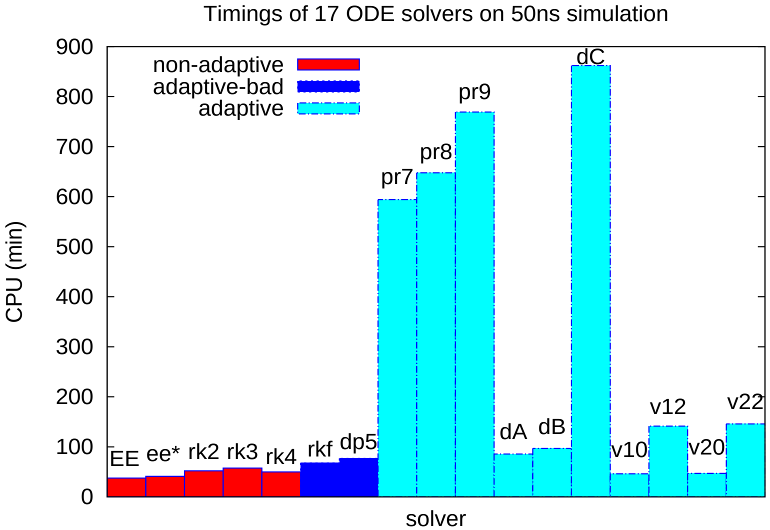
All solvers produce identical-looking plots
except RKFB4 and DoPri5 (their timings are shown in parentheses)

Comparison runs for $I_o = 12.e12\ W/m^2$, $\lambda = 532$ nm.

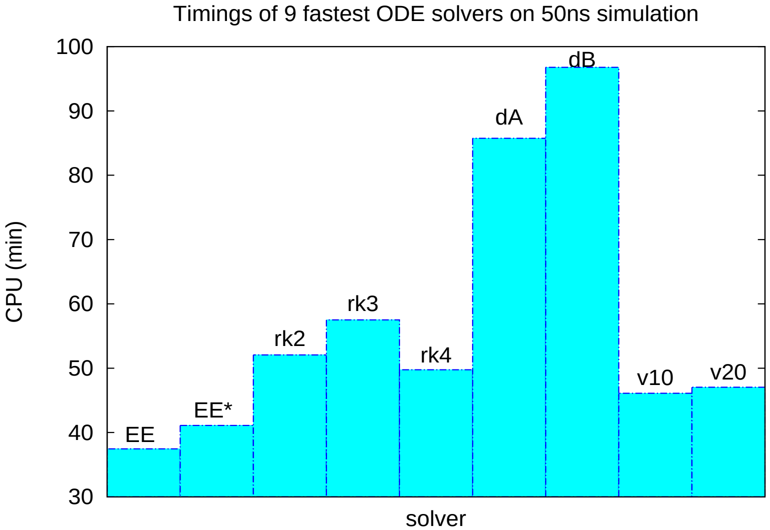
CPU timings of 17 ODE solvers for 50ns runs

time-stepper	factor	cpu (sec)	cpu (min)	comments
EE	50	2248	37	best!
EE	100	2466	41	
RK2	50	3124	52	
RK3	50	3451	58	
RK4	40	2985	50	
RKFB4	400	(4075)	(70)	inaccurate
DoPri5	500	(4610)	(77)	inaccurate
Peer 7	100	35643	594	
Peer 8	100	38869	648	
Peer 9	100	46143	769	
dode a	100	5144	86	
dode b	100	5806	97	
dode c	100	51717	862	worst
VODE 10	10	2766	46	2nd best
VODE 12	10	8490	142	
VODE 20	10	2823	47	3rd best
VODE 22	10	8748	146	

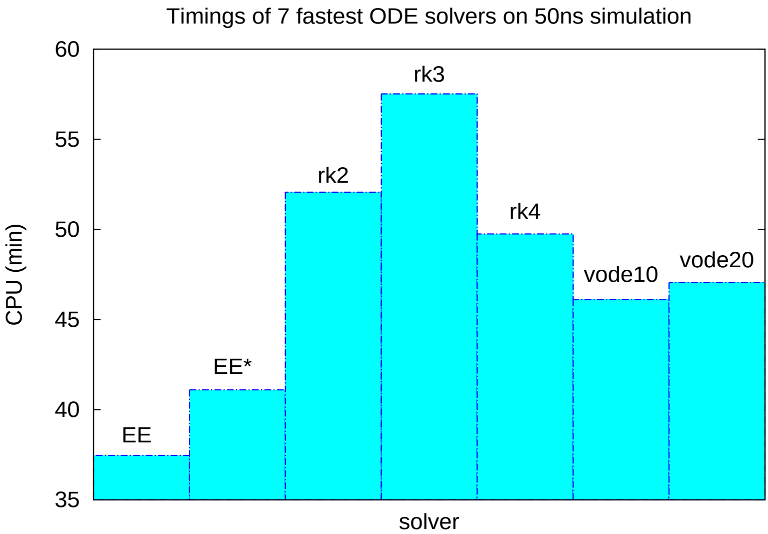
Timings of 17 ODE sovers on 50ns simulation



Timings of 9 best ODE sovers on 50ns simulation



Timings of the 7 best ODE sovers on 50ns simulation



Conclusions on time-steppers

- All solvers produce identical plots (agree by eye-norm) **except** rkfb4 and DoPri5: had to relax TOL and their plots are off.
- Lower order methods are much faster than higher oder (cost of RHS evaluation is very high, the fewer the better!)
- Non-adaptive methods performed better than adaptive !!!
- Stiff methods performed **worse** than non-stiff,
  indicating our ODE system is not as stiff as we thought...
- None could beat Explicit Euler !!!
  ...contrary to what numerical methods books usually preach...

The undisputed winner is Explicit Euler !
next come vode10 (24% slower), vode20 (27% slower),
rk4 (35% slower).

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- Intel Ordinary Differential Equations Solver Library http://software.intel.com/en-us/articles/intel-ordinary-differential-equ
- VODE_F90 ODE solver, Fortran 90 version of VODE ode solver by Brown, Byrne, and Hindmarsh. http://www.radford.edu/ thompson/vodef90web/

VODE method_flags

We use VODE with Method_Flag $= 10, 12, 20, 22$, meaning:

- METH $= 1$: implicit Adams method
- METH $= 2$: method based on BDF
- MITER $= 0$: functional iteration (no Jacobian involved).
- MITER $= 2$: chord iteration with internally generated (difference quotient) full Jacobian (using NEQ extra calls to F per df/dy value).

Reaction Rate Coefficients 1

Spontaneous Decay: $Cu_j \to Cu_i + \hbar\omega_{ij}$

- $$A_{ij} = 1.499 \times 10^{-8} \lambda_{ij}^2 \left(\frac{g_j}{g_i}\right) / f_{ij}, \;\; \nu_{\mathrm{dec},ij} = 10^8 A_{ji}$$

- i : lower effective level, j : upper effective level
  λ_{ij}: effective light wavelength (Å)
  A_{ij}: rate coefficient ($10^8 s^{-1}$)

Reaction Rate Coefficients 2

Collisional excitation/de-excitation: $Cu_i + e \leftrightarrow Cu_j + e$

- excitation rate coefficient

$$k_{exc,ij} = \frac{1.578 \times 10^{-3} \bar{g}\,(\Delta E_{ij}/T_e)\, e^{-\Delta E_{ij}/T_e} f_{ij}}{\Delta E_{ij}\sqrt{T_e}}\,(cm^3 s^{-1})$$

$$\bar{g}(y) = A + De^y \int_1^\infty t^{-1} e^{-(ty)}\, dt$$

- de-excitation rate coefficient

$$k_{dexc,ji} = k_{exc,ij}\left(\frac{g_i}{g_j}\right) e^{\Delta E_{ij}}$$

Reaction Rate Coefficients 3

Collisional ionization/three particle recombination

- Electron impact ionization

$$a_1 = 4 \times 10^{-14} \, (cm^2)$$

$$\sigma_{ei,i} = a_1 q_i \left(\frac{\ln(E/P_i)}{EP_i} \right), \; k_{ei} = 6.7 \times 10^7 \left(\frac{a_1 q_i}{T_e^{\frac{3}{2}}} \right) \frac{T}{P_i} E_1(P_i/T)$$

- three body recombination rate coefficient

$$k_{eq} = C_{\text{saha}} T_e^{\frac{3}{2}} (g_{Cu^+}/g_i) e^{-(IP_i/T_e)} \quad (cm^{-3})$$

$$k_{tbod,i} = k_{ei,i}/k_{eq} \times N_e \; (cm^3 s^{-1}), \; C_{\text{saha}} = 6 \times 10^{27}.$$

Reaction Rate Coefficients 4

Laser-induced photo ionization: $Cu_i + m\hbar\omega_{\text{laser}} \rightarrow Cu^+ + e$

- MPI rate for excited state

$$m_i = \mathrm{int}(\frac{IP_i}{\hbar\omega}+1),\ W_i = \left(\frac{\sigma_i}{\hbar\omega}\right)^{m_i}\frac{1}{\nu^{m_i-1}(m_i-1)!},(W/cm^2)^{-m_i}s^{-1}$$

$$\nu_{\text{phot}} = W_i \times I_{\text{las}}^{m_i}\ s^{-1}$$

- PI cross section excited state

$$\sigma_{vn}(cm^2) = 4.12^{26}\frac{I_n{}^{\frac{5}{2}}}{Z_n}\left(\frac{1}{\nu^3}\right)$$

The End

THANK YOU !