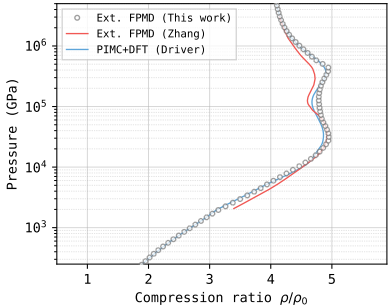
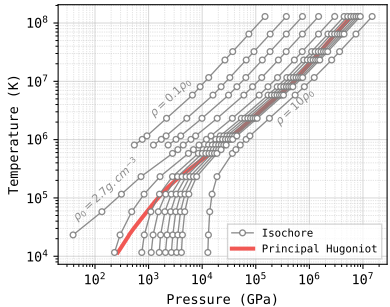
Extended FPMD model for high temperature simulations in ABINIT

Implementation and applications to hot aluminum and boron





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Usual plane wave approach at high temperature

We want to compute physical quantities in extreme temperature conditions as precisely as possible.

- Energy
- Entropy
- Pressure
- Electrical conductivity
- Conductivity
- Absorption/Reflectivity...

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Ab initio QMD methods most appropriate to get these quantities with high accuracy.

Electron density in plane wave DFT software

$$n(\mathbf{r}) = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \sum_{n=0}^{\infty} f_{\mathbf{k},n} \left| \Psi_{\mathbf{k},n}(\mathbf{r}) \right|^2 ,$$

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Kubo-Greenwood formulation of conductivity[1,2] (real part)

$$\sigma_1(\nu) = \frac{2\pi}{\Omega} \sum_{n}^{\infty} \sum_{m}^{\infty} \frac{\Delta f_{mn}}{\Delta \epsilon_{mn}} \langle \mathbf{k}, n | \nabla | \mathbf{k}, m \rangle \langle \mathbf{k}, m | \nabla | \mathbf{k}, n \rangle \delta(\Delta \epsilon_{mn} - \nu).$$

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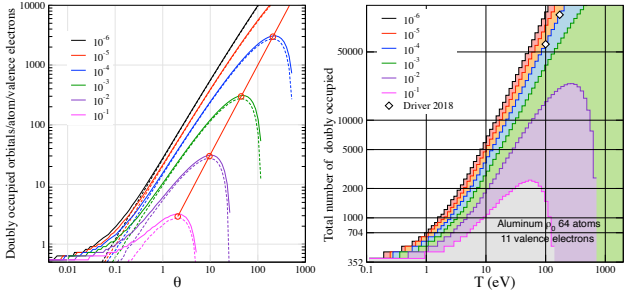
More and more bands needed at high temperature.

[1] R Kubo. Journal of the Physical Society of Japan, 12(6):570-586, 1957.

[2] D A Greenwood. Proceedings of the Physical Society, 71(4):585, 1958.

Last Level Occupancy (LLO) around 10^{-5} to ensure precise results.
$\Longrightarrow$ More than 50,000 bands at 100 eV for 64 Al at 2.7 g/cc.

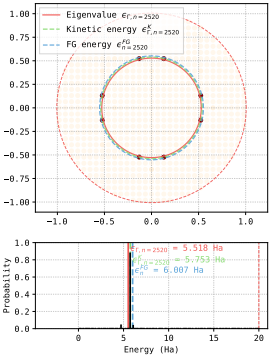


On the left: Universal curve giving the number of double occupied orbitals per atom and per valence electron in the HEG model vs degeneracy $\theta = T/T_F$. On the right: Number of orbitals needed to ensure fixed LLO vs temperature for a system of 64 aluminum atoms at standard density[3].

[3] A Blanchet, M Torrent, J Clérouin. Physics of Plasmas, 27(12):122706, 2020.

Extended FPMD model introduction

- High energy orbitals = pure PW.
- Setting high PW $\Longrightarrow$ Involves doing a full diagonalisation work of a big nearly empty matrix.
- Continuum at high energy $\Longrightarrow$ Split the quantities of interest into two contributions.



Plane wave coefficients of a high energy wave function on the reciprocal space plane $k_z = 0$. Dominant coefficients are represented with dark red opaque color. Bottom plot shows the square of these coefficients vs plane waves energy (integrated over all reciprocal space) for FCC aluminum system at $T = 20\,\text{eV}$[5].

[4] S Zhang, H Wang, W Kang, P Zhang, and X T He. Physics of Plasmas, 23(4):042707, 2016.

[5] A Blanchet, J Clérouin, M Torrent, F Soubiran. Upcoming paper on Extended FPMD model, 2021.

Restricted WF shape by imposing condition on the PW coefficients

$$C_{\mathbf{k},n}(\mathbf{G}) = C_{0_{\mathbf{k},n}} e^{\frac{-\left(\sqrt{2\epsilon_n^{FG}} - |\mathbf{k}+\mathbf{G}|\right)^2}{4\sigma^2}}.$$

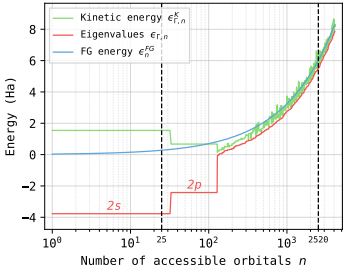
Contributions derived

$$N_e^{FG} = \frac{\sqrt{2}}{\pi^2}\frac{\Omega}{\beta^{3/2}}\mathcal{F}_{\frac{1}{2}}(\gamma, \beta\epsilon_{N_c}^{FG}),$$

$$E_K^{FG} = \frac{\sqrt{2}}{\pi^2}\frac{\Omega}{\beta^{5/2}}\mathcal{F}_{\frac{3}{2}}(\gamma, \beta\epsilon_{N_c}^{FG}).$$

$\Longrightarrow$ Electronic density, chemical potential, entropy and stress tensor.

No numerical unbounded integration required.



Energies versus number of accessible doubly occupied otbitals for a 32 aluminum atoms system at $\mathbf{k} = \Gamma$ point at $T = 20$ eV and standard density. Eigenvalues shown in red, Kinetic energy in green and Fermi gas (FG) energy in blue[5].

New module, *m_extfpmd* at the same level as *m_occ* (61) and new type, *extfpmd_type*.

Implementation

- Initialization in *m_gstate* before the SCF cycle, contribution computed in *m_vtorho*.
- Can start with input WF file.
- Contributions are printed in the output for post-treatment.

Usage

- One-parameter activated (*use_extfpmd 1*).
- Reduce drastically the number of bands.
- Warning is sent if model error δ^{FG} is over a predefined threshold inviting you to raise *nband*.
- Use small core or All-electrons PSP with SC radius.
- Cutoff convergency is still needed.

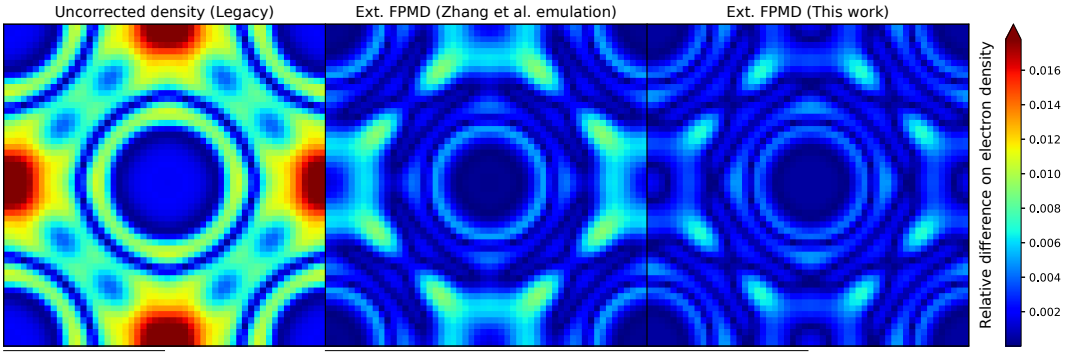
m_extfpmd module header in ABINIT.

```
!!****t* m_extfpmd/extfpmd_type
!! NAME
!! extfpmd_type
!!
!! FUNCTION
!! Store extfpmd functions and parameters.
!!
!! SOURCE
type,public :: extfpmd_type
  integer :: bcut,nbcut,nfftf,nspden,version
  real(dp) :: e_bcut,edc_kinetic,e_kinetic,entropy
  real(dp) :: nelect,shiftfactor,ucvol
  real(dp),allocatable :: vtrial(:,:)
contains
  procedure :: compute_e_kinetic
  procedure :: compute_entropy
  procedure :: compute_nelect
  procedure :: compute_shiftfactor
  procedure :: init
  procedure :: destroy
end type extfpmd_type
!!***
```

Ext. FPMD model elementary tests

- Electron density correction (for FCC Aluminum, T=20 eV)

Relative error[6] on the electron density on the plane $z = 0$ with only 32 bands per atom.



[6] Reference computed with 512 bands per atom (LLO < E-16).

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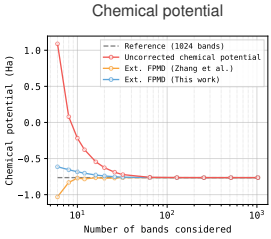
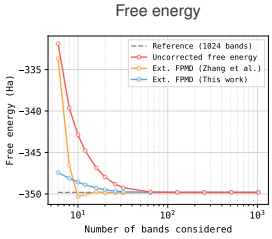
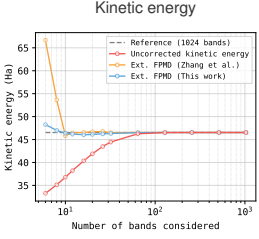
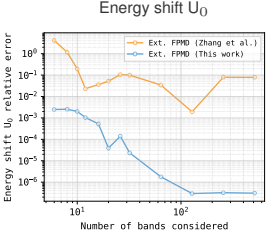
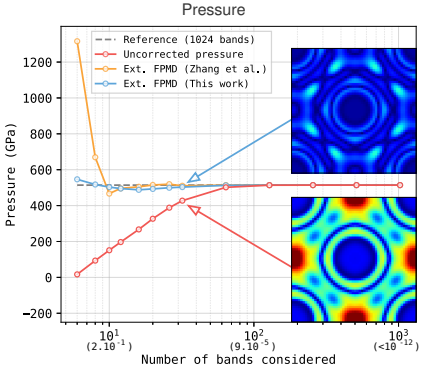




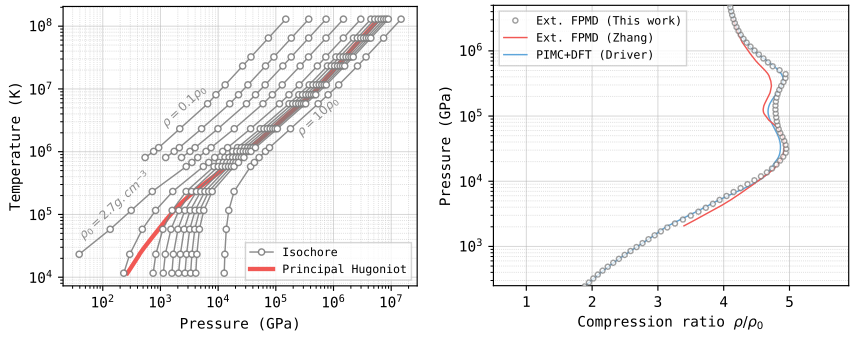






- Using EOS codes (FPEOS, SESAME)[7].
- Aluminum ($Z = 13$): 260 full MD, from $T = 1$ eV to $T = 11$ keV ($0.1\rho_0 < \rho_0 < 10\rho_0$).

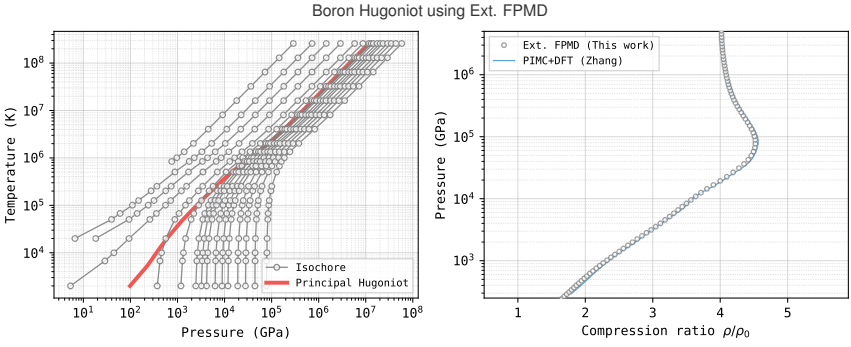
Aluminum Hugoniot using Ext. FPMD[5]



[7] B Militzer, F Gonzalez-Cataldo, S Zhang, K P Driver, F Soubiran. Physical Review E 103:013203, 2021.

Hugoniot equation of state curves

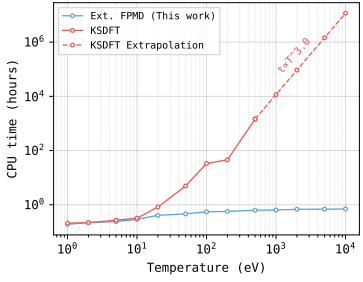
- Boron ($Z = 5$): 384 full MD.
- Temperatures goes from $T = 300\,\mathrm{K}$ to $T = 44\,\mathrm{keV}$.
- Compression goes from $\rho = 0.25\,\mathrm{g/cc}$ ($0.1\rho_0$) to $\rho = 50\,\mathrm{g/cc}$ ($20\rho_0$).

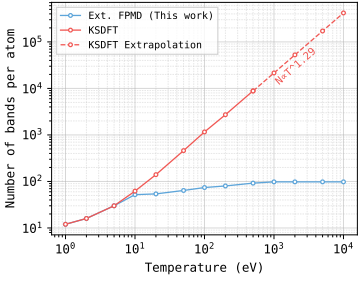


Speedup evaluation

- Speedup tests on Broadwell Intel processors.
- The standard KSDFT shows a T^3 dependency.
- Ext. FPMD weakly depends on the temperature.
- Much less bands are used in Ext. DFT. At 200 eV, KSDFT uses 2,720 bands to fullfill a LLO of about 10^{-5}. Corresponding Ext. FPMD uses 80 bands.

Evolution of CPU time with temperature of FCC aluminum.





Conclusion

- Efficiency of the Ext. FPMD method
  - Easy to use (single variable activation).
  - Lot of computing time saved.
  - Estimate of error due to band cut and Warning if things goes wrong.
- Advantages compared to Zhang et al. implementation
  - Open source (Merged in ABINIT v9.5.2).
  - No buffer to evaluate the shift factor U_0.
  - Warning send if error too high.
  - No unbounded numerical integrals done (fast for high temperature).

Thank you for your attention!

(1) R Kubo. Journal of the Physical Society of Japan, 12(6):570-586, 1957.
(2) D A Greenwood. Proceedings of the Physical Society, 71(4):585, 1958.
(3) A Blanchet, M Torrent, J Clérouin. Physics of Plasmas, 27(12):122706, 2020.
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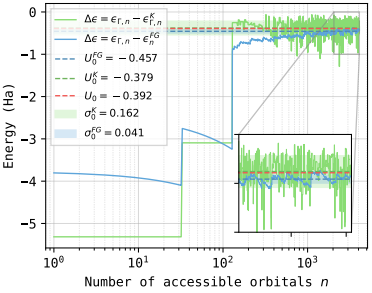
(5) A Blanchet, J Clérouin, M Torrent, F Soubiran. Upcoming paper on Extended FPMD model, 2021.
(7) B Militzer, F Gonzalez-Cataldo, S Zhang, K P Driver, F Soubiran. Physical Review E 103:013203, 2021.

Fermi Gas energy $\epsilon_n^{FG} = \frac{1}{2}\left(6\pi^2 n\right)^{2/3},$

Model error $\delta^{FG} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \left| \frac{\epsilon_{\mathbf{k},N_c} - \epsilon_{N_c}^{FG} - U_0}{\epsilon_{\mathbf{k},N_c}} \right|.$



Difference between the kinetic energy $\epsilon^K_{\Gamma,n}$ and eigenvalues $\epsilon_{\Gamma,n}$ is shown in solid line green color. The estimation of the energy shift factor U_0^K using the kinetic energy computed with the lasts 1000 bands is plotted in dashed horizontal green line, and the attached standard deviation σ_0^K is represented by the green area near the U_0^K constant line. The same quantities computed with the Fermi gas model are plotted in blue.