

# Implementation of the Strain Perturbation in ABINIT

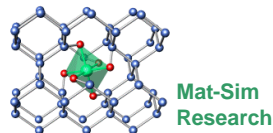
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Supported by The Center for Piezoelectrics by Design, ONR grant N00014-01-1-0365 and Mat-Sim Research, [www.mat-simresearch.com](http://www.mat-simresearch.com)



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# Overview

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- *Goal:* Direct calculation of the elastic and piezoelectric tensors and related quantities
- Strain and the Abinit reduced-coordinate formulation
- Brief review of density functional perturbation theory
- Response function (RF) code organization
- Development process design
- Special issues: nonlocal pseudopotentials, symmetry, non-linear core corrections, cutoff smoothing, metals
- Using: new input and output
- Atom coordinate relaxation and anaddb post processing
- Future development issues
- (Appendix)



# Strain tensor $\eta_{\alpha\beta}$ as a perturbation

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- Strain really only changes the positions of the atomic (pseudo)potentials,

$$V_{ext}(\mathbf{r}) = \sum_{\mathbf{R}} \sum_{\tau}^{\text{cell}} V_{\tau}(\mathbf{r} - \boldsymbol{\tau} - \mathbf{R}) \xrightarrow{\eta} V_{ext}(\mathbf{r}) = \sum_{\mathbf{R}} \sum_{\tau}^{\text{cell}} V_{\tau}[\mathbf{r} - (\mathbf{1} + \boldsymbol{\eta}) \cdot \boldsymbol{\tau} - (\mathbf{1} + \boldsymbol{\eta}) \cdot \mathbf{R}].$$

- However, this causes unique problems for perturbations expansions:
  - Viewed in terms of the infinite lattice, the strain perturbation can never be small.
  - From the point of view of a single unit cell, strain changes the periodic boundary conditions, so wave functions of the strained lattice cannot be expanded in terms of those of the unstrained lattice.
- Strain appears to be qualitatively different from other perturbations such as periodicity-preserving atomic displacements



# Strain tensor $\eta_{\alpha\beta}$ as a perturbation, continued

- Existing formulation<sup>(1)</sup>
  - Introduce fictitious strained self-consistent Hamiltonian obtained by a scale transformation of the unstrained Hamiltonian,
$$\tilde{H}_{SCF}^{\eta}(\mathbf{r}, \nabla) = H_{SCF} \left[ (1 + \boldsymbol{\eta})^{-1} \cdot \mathbf{r}, (1 + \boldsymbol{\eta}) \cdot \nabla \right]$$
  - $\tilde{H}_{SCF}^{\eta}$  obeys the same boundary conditions as the strained Hamiltonian
  - $\tilde{H}_{SCF}^{\eta}$  has the same spectrum unstrained Hamiltonian, and a simply related electron density  $\tilde{n}^{\eta}(\mathbf{r})$
  - Energy difference between unstrained crystal and fictitious strained crystal described by  $\tilde{H}_{SCF}^{\eta}$  is easily obtained
- Problem: Hartree and XC potentials of  $\tilde{H}_{SCF}^{\eta}$  are not those generated by  $\tilde{n}^{\eta}(\mathbf{r})$  so  $\tilde{H}_{SCF}^{\eta}$  is not a genuine Kohn-Sham Hamiltonian
  - Must introduce modified external potential  $\tilde{V}_{ext}^{\eta}$  to compensate
  - Then use DFPT treating  $V_{ext}^{\eta} - \tilde{V}_{ext}^{\eta}$  as the perturbation
- Complex two-step process apparently changes the structure of DFPT from that for other perturbations



# Abinit reduced coordinate ( $\sim$ ) formulation

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- Every lattice, unstrained or strained, is a unit cube in reduced coordinates.
  - Primitive real and reciprocal lattice vectors define the transformations:

$$\mathbf{X}_\alpha = \sum_i R_{\alpha i}^{\text{P}} \tilde{\mathbf{X}}_i, \quad \mathbf{K}_\alpha \equiv (\mathbf{k}_\alpha + \mathbf{G}_\alpha) = \sum_i G_{\alpha i}^{\text{P}} \tilde{\mathbf{K}}_i, \quad \sum_\alpha R_{\alpha i}^{\text{P}} G_{\alpha j}^{\text{P}} = 2\pi\delta_{ij}$$

- Cartesian indices  $\alpha, \beta, \dots = 1, 3$  and reduced indices  $i, j, \dots = 1, 3$
- **Every term in the DFT functional can be expressed in terms of dot products and the unit cell volume  $\Omega$ .**
  - Dot products and  $\Omega$  in reduced coordinates are computed with **metric tensors**,

$$\mathbf{X}' \cdot \mathbf{X} = \sum_{ij} \tilde{\mathbf{X}}'_i \boldsymbol{\Xi}_{ij} \tilde{\mathbf{X}}_j, \quad \mathbf{K}' \cdot \mathbf{K} = \sum_{ij} \tilde{\mathbf{K}}'_i \boldsymbol{\Upsilon}_{ij} \tilde{\mathbf{K}}_j, \quad \Omega = (\det[\boldsymbol{\Xi}_{ij}])^{1/2}$$

- Strain is now a “simple” parameter of a density functional whose wave functions have invariant boundary conditions.



# Abinit reduced coordinate ( $\sim$ ) formulation

- Strain derivatives act **only** on the metric tensors,

$$\Xi_{ij}^{(\alpha\beta)} \equiv \frac{\partial \Xi_{ij}}{\partial \eta_{\alpha\beta}} = R_{\alpha i}^P R_{\beta j}^P + R_{\beta i}^P R_{\alpha j}^P, \quad \Upsilon_{ij}^{(\alpha\beta)} \equiv \frac{\partial \Upsilon_{ij}}{\partial \eta_{\alpha\beta}} = -G_{\alpha i}^P G_{\beta j}^P - G_{\beta i}^P G_{\alpha j}^P$$

$$\begin{aligned} \Xi_{ij}^{(\alpha\beta\gamma\delta)} \equiv \frac{\partial^2 \Xi_{ij}}{\partial \eta_{\gamma\delta} \partial \eta_{\alpha\beta}} &= \delta_{\alpha\gamma} (R_{\beta i}^P R_{\delta j}^P + R_{\delta i}^P R_{\beta j}^P) + \delta_{\beta\gamma} (R_{\alpha i}^P R_{\delta j}^P + R_{\delta i}^P R_{\alpha j}^P) \\ &+ \delta_{\alpha\delta} (R_{\beta i}^P R_{\gamma j}^P + R_{\gamma i}^P R_{\beta j}^P) + \delta_{\beta\delta} (R_{\alpha i}^P R_{\gamma j}^P + R_{\gamma i}^P R_{\alpha j}^P), \end{aligned}$$

- $\Omega$  has uniquely simple derivatives for **Cartesian Strains**

$$\frac{\partial \Omega}{\partial \eta_{\alpha\beta}} = \delta_{\alpha\beta} \Omega, \quad \frac{\partial^2 \Omega}{\partial \eta_{\alpha\beta} \partial \eta_{\gamma\delta}} = \delta_{\alpha\beta} \delta_{\gamma\delta} \Omega$$

- **Key decision:** strain will be Cartesian throughout the code
  - Existing perturbations will remain in reduced-coordinates



# Stress and strain notation

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Cartesian	xx	yy	zz	yz	xz	xy
Cartesian	1 1	2 2	3 3	2 3	1 3	1 2
Voigt	1	2	3	4	5	6
ipert, idir	natom+3, 1	natom+3, 2	natom+3, 3	natom+4, 1	natom+4, 2	natom+4, 3

- All forms used at various places internally and in output



# Density Functional Perturbation Theory

- All quantities are expanded in power series in a DF energy parameter  $\lambda$ ,

$$X(\lambda) = X^{(0)} + \lambda X^{(1)} + \lambda^2 X^{(2)} + \dots, \quad X = E_{el}, T, V_{ext}, \psi_\alpha(\mathbf{r}), n(\mathbf{r}), \varepsilon_\alpha, H$$

- Solutions  $\psi^{(0)}$  of Kohn-Sham equation minimize the usual DFT functional  $E^{(0)}$

$$H^{(0)} \left| \psi_\alpha^{(0)} \right\rangle = \varepsilon_\alpha^{(0)} \left| \psi_\alpha^{(0)} \right\rangle.$$

- The variational functional for  $E^{(2)}$  is minimized by solutions  $\psi^{(1)}$  of the self-consistent Sternheimer equation

$$P_c (H^{(0)} - \varepsilon_\alpha^{(0)}) P_c \left| \psi_\alpha^{(1)} \right\rangle = -P_c H^{(1)} \left| \psi_\alpha^{(0)} \right\rangle,$$

- where  $P_c$  is the projector on unoccupied states (conduction bands) and

$$H^{(1)} = T^{(1)} + V_{ext}^{(1)} + V_{Hxc}^{(1)}, \quad V_{Hxc}^{(1)} = \left. \frac{\partial}{\partial \lambda} \frac{\delta E_{Hxc}}{\delta n(\mathbf{r})} \right|_{n^{(0)}} + \int \frac{\delta^2 E_{Hxc}}{\delta n(\mathbf{r}) \delta n(\mathbf{r}')} n^{(1)}(\mathbf{r}') d\mathbf{r}',$$

$$n^{(1)}(\mathbf{r}) = \sum_{\alpha}^{occ} [\psi_\alpha^{*(1)}(\mathbf{r}) \psi_\alpha^{(0)}(\mathbf{r}) + \psi_\alpha^{*(0)}(\mathbf{r}) \psi_\alpha^{(1)}(\mathbf{r})].$$



# DFPT for elastic and piezoelectric tensors

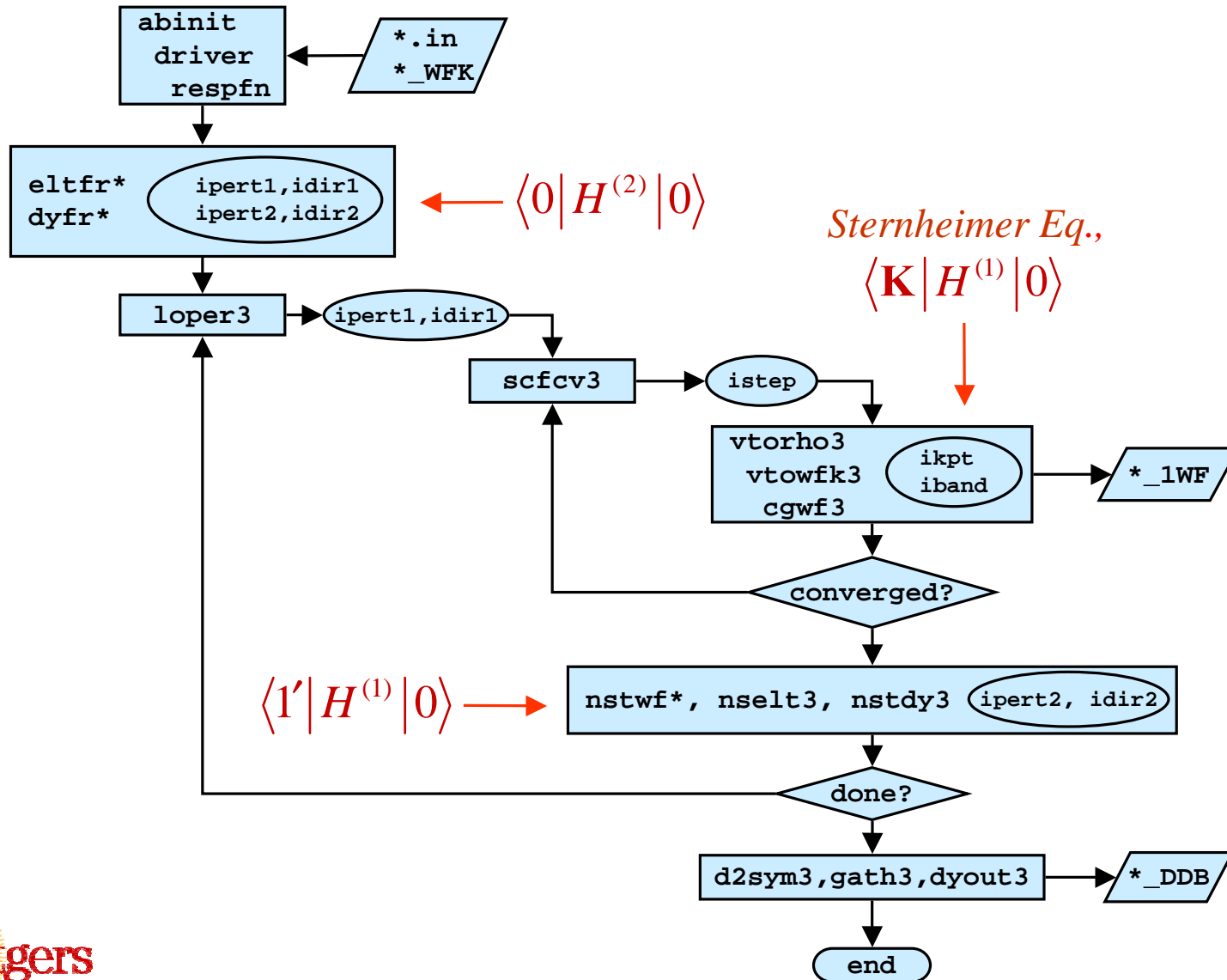
- Mixed 2<sup>nd</sup> derivatives of the energy with respect to two perturbations are needed.
  - By the “2n+1” theorem, these only require one set of 1<sup>st</sup> order wave functions,

$$E_{el}^{(\lambda_1\lambda_2)} = \sum_{\alpha}^{\text{occ}} \langle \psi_{\alpha}^{(\lambda_2)} | (T^{(\lambda_1)} + V_{ext}^{(\lambda_1)} + H_{Hxc0}^{(\lambda_1)}) | \psi_{\alpha}^{(0)} \rangle + \sum_{\alpha}^{\text{occ}} \langle \psi_{\alpha}^{(0)} | (T^{(\lambda_1\lambda_2)} + V_{ext}^{(\lambda_1\lambda_2)}) | \psi_{\alpha}^{(0)} \rangle + \frac{1}{2} \frac{\partial^2 E_{Hxc}}{\partial \lambda_1 \partial \lambda_2} \Big|_{n^{(0)}} ,$$

- Including atomic relaxation, we need
  - Clamped-atom elastic tensor -----  $\partial^2 E_{el} / \partial \eta_{\alpha\beta} \partial \eta_{\gamma\delta}$
  - Internal strain tensor -----  $\partial^2 E_{el} / \partial \eta_{\alpha\beta} \partial \tilde{\tau}_j$
  - Interatomic force constants -----  $\partial^2 E_{el} / \partial \tilde{\tau}_i \partial \tilde{\tau}_j$  ← Available
  - Clamped-atom piezoelectric tensor ----  $\partial^2 E_{el} / \partial \eta_{\alpha\beta} \partial \tilde{\mathcal{E}}_j$
  - Born effective charges -----  $\partial^2 E_{el} / \partial \tilde{\tau}_i \partial \tilde{\mathcal{E}}_j$  ← Available



# Response function code organization



# Design of development process

- Four stages based on RF code organization and degree of complexity
  - First  $\langle \mathbf{K} | H^{(1)} | 0 \rangle$ , second *Sternheimer*, third  $\langle 0 | H^{(2)} | 0 \rangle$ , and fourth  $\langle 1' | H^{(1)} | 0 \rangle$
- **Stage-by-stage and term-by-term validation based on existing GS first derivatives of the total energy (1DTE's)**
- First stage
  - Re-compute stress as  $\langle 0 | H^{(1)} | 0 \rangle = \sum_{\mathbf{K}} \langle 0 | \mathbf{K} \rangle \langle \mathbf{K} | H^{(1)} | 0 \rangle$
  - Compare term-by-term to stress breakdown available in GS calculation:

```
stress: component      1 of hartree stress is  -8.625241635590E-04
stress: component      2 of hartree stress is  -7.368896556922E-04
stress: component      1 of loc psp stress is   2.656792257661E-03
stress: component      2 of loc psp stress is   2.166978270656E-03
stress: component      1 of xc stress is       4.902613744139E-03
stress: component      2 of xc stress is       4.902613744139E-03
stress: ii (diagonal) part is  -7.753477394392E-03
stress: component      1 of kinetic stress is  -5.477053634248E-03
stress: component      2 of kinetic stress is  -5.272489903492E-03
stress: component      1 of nonlocal ps stress is  2.396391029219E-03
stress: component      2 of nonlocal ps stress is  1.907769135572E-03
stress: component      1 of Ewald energy stress is  5.707853334522E-03
stress: component      2 of Ewald energy stress is  6.846039389167E-03
stress: component      1 of core xc stress is  -1.789801472019E-03
stress: component      2 of core xc stress is  -1.911589314962E-03
```



# Design of development process (continued)

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- Second through fourth stage – numerical derivatives of GS quantities
  - “Five-point” strain first derivatives of GS quantities (symmetric shear strains)
  - Strain increments small enough to keep complete  $\{\mathbf{K}\}$  set invariant
  - Second derivatives of total energy (2DTE’s) from numerical derivatives of 1DTE’s (eg., stress)
  - **Extreme** convergence of self-consistency required, but not of  $\mathbf{k}$ ’s or cutoffs
- Second stage – *Sternheimer*
  - Convergence of self-consistency loop
  - Ensure variational part of RF 2DTE’s decreases with convergence
  - First-order density  $n^{(1)}$  validated by comparison with numerical  $n^{(0)}$  derivatives
- Second stage – validation of variational RF 2DTE’s
  - Non-variational part  $\langle 0|H^{(2)}|0\rangle$  not yet available
  - Compute numerical 2DTE’s as above with converged GS wave functions for strained lattices
  - Subtract numerical 2DTE’s with strained lattices but unstrained (“frozen”) GS wave functions
- So far, we are only dealing with “diagonal” 2DTE’s



# Design of development process (continued)

- Third stage –  $\langle 0 | H^{(2)} | 0 \rangle$  :

$$E_{\text{non-var}}^{(\lambda_1 \lambda_2)} = \sum_{\alpha}^{\text{occ}} \langle \psi_{\alpha}^{(0)} | (T^{(\lambda_1 \lambda_2)} + V_{\text{ext}}^{(\lambda_1 \lambda_2)}) | \psi_{\alpha}^{(0)} \rangle + \frac{1}{2} \frac{\partial^2 E_{Hxc}}{\partial \lambda_1 \partial \lambda_2} \Big|_{n^{(0)}}$$

- Validate term-by-term with “frozen wave function” numerical strain derivatives of stress components
- Diagonal and off-diagonal 2DTE’s
- Numerical strain derivatives of forces for internal strain mixed 2DTE’s
- Frozen wf strain derivatives of the electric polarization are zero, so there is no contribution to piezoelectric tensor from these terms
- Note that the numerical derivatives of stress  $\sigma$  need  $\Omega$  factor for 2DTE comparison

$$\frac{\partial^2 E_{el}}{\partial \eta_{\alpha\beta} \partial \eta_{\gamma\delta}} = \frac{\partial}{\partial \eta_{\alpha\beta}} (\Omega \sigma_{\gamma\delta})$$



# Design of development process (continued)

- Fourth stage – 
$$E_{\text{non-stat}}^{(\lambda_1 \lambda_2)} = \sum_{\alpha}^{\text{occ}} \left\langle \psi_{\alpha}^{(\lambda_2)} \left| (T^{(\lambda_1)} + V_{\text{ext}}^{(\lambda_1)} + H_{Hxc0}^{(\lambda_1)}) \right| \psi_{\alpha}^{(0)} \right\rangle$$
  - Use strain-perturbation wave functions  $\psi_{\alpha}^{(\lambda_2)}$
  - $\lambda_1$  is Cartesian strain or reduced atomic displacement
- Electric field is special case – 
$$\frac{\partial^2 E_{el}}{\partial \tilde{\mathcal{E}}_j \partial \eta_{\alpha\beta}} = \frac{2\Omega}{(2\pi)^3} \int_{\text{BZ}} \sum_m^{\text{occ}} \left\langle i \psi_{\mathbf{k}m}^{(\tilde{k}_j)} \left| \psi_{\mathbf{k}m}^{(\eta_{\alpha\beta})} \right\rangle d\mathbf{k}$$
  - Field and  $d/dk$  first-order wave function in reduced coordinates
- Validate using numerical strain derivatives of GS stresses, forces, and polarization
  - Use converged strained wave functions
  - No subtractions since non-variational contributions are already validated
  - For polarization, numerical derivatives have to be corrected to give “proper” piezoelectric tensor (see `Infos/theory/1r.pdf`)
  - Need to use finite-difference  $d/dk$  wf’s in RF calculation for accurate comparison

Non-self-consistent



# Subroutines for strain perturbation (53)

## Modified routines

	K10	1'10	020	Utl
cart29.f				•
cgwf3.f	•			
chkinp.f				•
dyout3.f				•
eneres3.f	•			
gath3.f				•
insy3.f	•	•		
loper3.f	•	•		
mkcor3.f	•	•	•	
mkffkg3.f	•	•	•	
mkffkg.f	•	•	•	
mkffnl.f	•	•	•	
nonlop.f	•	•	•	
nstdy3.f		•		
opernl2.f	•	•	•	
opernl3.f	•	•	•	
opernl4a.f	•	•	•	
opernl4b.f	•	•	•	
prtene3.f	•			
prtxf.f				•
respfn.f	•	•	•	
scfcv3.f	•			
vtorho3.f	•			
vtowfk3.f	•			

## New routines

	K10	1'10	020	Utl
contistr01.f			•	
contistr03.f			•	
contistr10.f			•	
contistr12.f			•	
contistr21.f			•	
contistr30.f			•	
contstr21.f			•	
contstr22.f			•	
contstr23.f			•	
contstr24.f			•	
contstr25a.f			•	
contstr25.f			•	
contstr26.f			•	

	K10	1'10	020	Utl
d2kindstr2.f			•	
eltfrhar3.f			•	
eltfrkin3.f			•	
eltfrloc3.f			•	
eltfrnl3.f			•	
eltfrxc3.f			•	
ewald4.f			•	
hartrestr.f	•	•		
kpgstr.f	•	•		
metstr.f	•	•		
newfermiel.f	•			
nselt3.f		•		
nstwf4.f		•		
splfit2.f			•	
symkchk.f				•
vlocalstr.f	•	•		

K10
1'10
020
Utl

$\langle \mathbf{K} | H^{(1)} | 0 \rangle$ 
 $\langle 1' | H^{(1)} | 0 \rangle$ 
 $\langle 0 | H^{(2)} | 0 \rangle$ 
*Utility*

Mnemonics

**str** – strain, **eltfr** – elastic tensor frozen  
**istr** – internal strain, **cont** – contraction

# Nonlocal pseudopotentials in Abinit<sup>1</sup>

- Most mathematically complex object for strain derivatives
- Reduced wave vector matrix elements have the form

$$\langle \tilde{\mathbf{K}}' | V_{NL} | \tilde{\mathbf{K}} \rangle = \frac{1}{\Omega} \sum_{\kappa\ell} e^{2\pi i \tilde{\mathbf{K}}' \cdot \tilde{\tau}_\kappa} f_{\kappa\ell} \left( \sum_{ij} \Upsilon_{ij} \tilde{K}'_i \tilde{K}'_j \right) \times \\ \wp_\ell \left( \sum_{ij} \Upsilon_{ij} \tilde{K}'_i \tilde{K}'_j, \sum_{ij} \Upsilon_{ij} \tilde{K}_i \tilde{K}_j, \sum_{ij} \Upsilon_{ij} \tilde{K}_i \tilde{K}_j \right) e^{-2\pi i \tilde{\mathbf{K}} \cdot \tilde{\tau}_\kappa} f_{\kappa\ell} \left( \sum_{ij} \Upsilon_{ij} \tilde{K}_i \tilde{K}_j \right)$$

- $\wp_\ell$  modified Legendre polynomials,  $f_{\kappa\ell}$  psp form factors,  $\tilde{\tau}_\kappa$  reduced atom coordinates
- All arguments are dot products expressed with metric tensors
- Psp's act on wave functions (in `open1*.f`) by summing wave function coefficients  $c_{\alpha\tilde{\mathbf{K}}}$  times a set of tensor products of reduced  $\tilde{\mathbf{K}}$  components,

$$T_{\ell m}(\tilde{\mathbf{K}}) = \tilde{K}_1^{I_T(1,\ell,m)} \tilde{K}_2^{I_T(2,\ell,m)} \tilde{K}_3^{\ell - I_T(1,\ell,m) - I_T(2,\ell,m)}$$

- $I_T(i, \ell, m)$  are index arrays
- $T_{\ell m}(\tilde{\mathbf{K}})$  (created in `mkfkg.f`) are *analogous* to spherical harmonics



# Nonlocal pseudopotential strain derivatives

- *Mathematica* programs created to do symbolic differentiation and extract coefficients coupling pairs of “input” ( $\tilde{\mathbf{K}}$ ) and “output” ( $\tilde{\mathbf{K}}'$ ) tensors
- Coefficients are polynomials in  $\Upsilon_{ij}, \Upsilon_{ij}^{(\alpha\beta)}, \Upsilon_{ij}^{(\gamma\delta)}, \Upsilon_{ij}^{(\alpha\beta\gamma\delta)}$
- SED and C programs turn *Mathematica* results into useful Fortran 90
  - Example from `contstr24.f`

```
cm(6,10)=(gm(1,3)**2*(270*dgm01(2,3)*dgm10(1,1)+540*dgm01(1,3)&
& *dgm10(1,2)+540*dgm01(1,2)*dgm10(1,3)+270*dgm01(1,1)*dgm10(2,3))&
& +gm(3,3)*(-108*gm(1,2)*(dgm01(1,3)*dgm10(1,1)+dgm01(1,1)*dgm10(1,3))&
& +gm(1,1)*(-54*dgm01(2,3)*dgm10(1,1)-108*dgm01(1,3)*dgm10(1,2)&
& -108*dgm01(1,2)*dgm10(1,3)-54*dgm01(1,1)*dgm10(2,3)))+gm(2,3)&
& *(540*gm(1,3)*(dgm01(1,3)*dgm10(1,1)+dgm01(1,1)*dgm10(1,3))-54*gm(1,1)&
& *(dgm01(3,3)*dgm10(1,1)+dgm01(1,1)*dgm10(3,3))+270*gm(1,3)**2*d2gm(1,1)&
& +gm(3,3)*(-108*dgm01(1,1)*dgm10(1,1)-54*gm(1,1)*d2gm(1,1)))+180*gm(1,3)&
& **3*d2gm(1,2)+gm(1,3)*(-108*(gm(1,2)*(dgm01(3,3)*dgm10(1,1)+dgm01(1,1)&
& *dgm10(3,3))+gm(1,1)*(dgm01(3,3)*dgm10(1,2)+dgm01(1,2)*dgm10(3,3)))&
& +gm(3,3)*(-216*dgm01(1,2)*dgm10(1,1)-216*dgm01(1,1)*dgm10(1,2)&
& -108*gm(1,2)*d2gm(1,1)-108*gm(1,1)*d2gm(1,2)))/36.d0
```

- Here  $gm(i,j)$ ,  $dgm*(i,j)$ ,  $d2gm(i,j)$  are  $\Upsilon_{ij}$ , etc.
- Many 1000's of lines of infrequently executed code in `cont*str*.f` and `metstr.f`
- Evaluation of `cm`'s is not a major factor in execution time



# Symmetry with the strain perturbation

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- The reduced-zone  $\mathbf{k}$  sample determined for (space group / strain) is used for  $\langle \mathbf{K} | H^{(1)} | 0 \rangle$ , *Sternheimer*, and  $\langle 1' | H^{(1)} | 0 \rangle$ 
  - The full-zone sample specified in input data must have the full space group symmetry (enforced by test).
- Loop on (`ipert1`, `idir1`) for 1<sup>st</sup>-order wave functions restricted by input variables (`rfstrs`, `rfdir`) but not by symmetry
  - This could be improved, but would have limited impact on performance
- Inner loop on (`ipert2`, `idir2`) in  $\langle 1' | H^{(1)} | 0 \rangle$  calculations is carried over all strain and atomic displacement terms
  - piezoelectric contribution is computed if  $d/dk$  wf's are available
- All  $\langle 0 | H^{(2)} | 0 \rangle$  strain and internal-strain tensor elements are computed, using the full zone  $\mathbf{k}$  sample
  - It is more efficient here to keep loops on strains and displacements inside routines like `nonlop.f`
  - The reduced zone for pairs of perturbations would seldom be reduced much anyway



# XC non-linear core correction

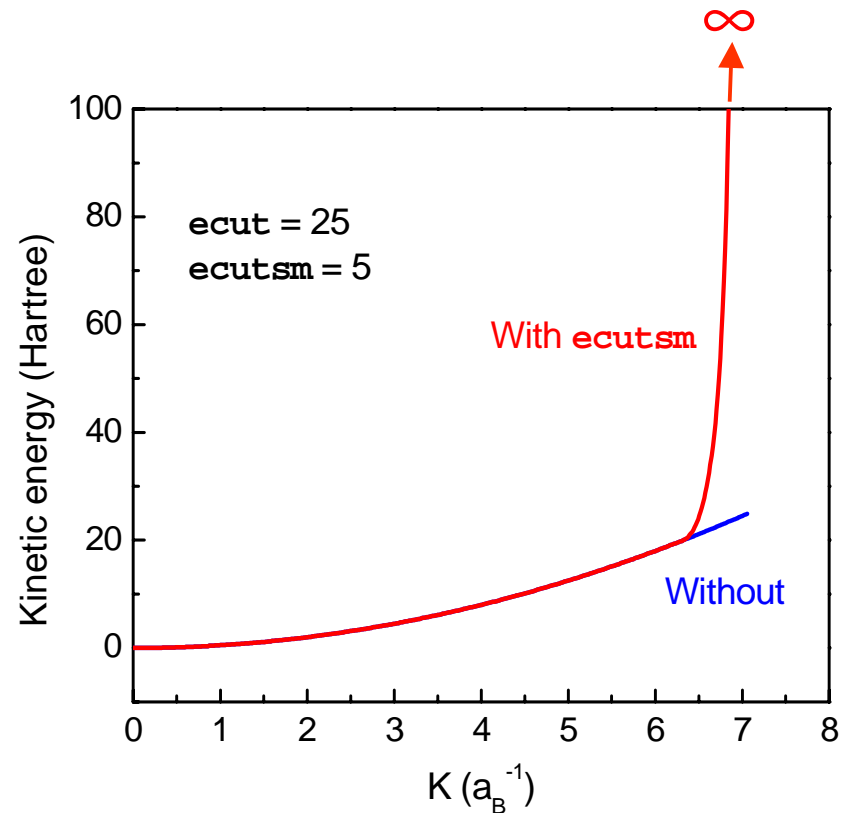
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- On the reduced real-space grid, electron charge depends only on  $\Omega^{-1}$
- Model core charge has a detailed dependence on  $E_{ij}$ 
  - Resulting analysis is rather complex
- Core charges must be **extremely smooth** functions to avoid significant convergence errors
  - Reason: Strain and atomic position derivatives of the xc self-interaction of a single core don't cancel point-by-point on the grid, but only in the integral
  - Inconsistencies in the treatment of the core charges and their derivatives in some `src_2psp/psp*cc.f` routines makes matters worse



# Kinetic energy cutoff “smoothing”

- Existing Abinit strategy to smooth energy dependence on lattice parameters in GS calculations
- RF strain derivative calculations do accurately reproduce GS numerical derivatives with non-zero `ecutsm`
- Divergence can produce large shifts in elastic tensor if calculation is not *very well* converged with respect to `ecut`
  - Remember, we take two derivatives
  - Perhaps the cutoff function could be improved



# Strain perturbation for metals

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- Thermal smearing of the Fermi surface must be introduced for stability
- In RF calculations, a band of partially-occupied states around  $\mathcal{E}_F$  is treated by finite-temperature perturbation theory in the Sternheimer solution, and only the completely unoccupied states are found by the conjugate-gradient method <sup>(1)</sup>
- For strain, a first-order Fermi energy  $\mathcal{E}_F^{(1)}$  must be introduced<sup>(2)</sup>
- $\mathcal{E}_F^{(1)}$  enters into the Sternheimer self-consistency process
- Convergence can be rather slow
  - Only simple mixing is presently used to iterate  $\mathcal{E}_F^{(1)}$
  - Coupling to the first-order potential iteration through Anderson or CG mixing may help
- Is  $\mathcal{E}_F^{(1)}$  needed for the  $\mathbf{Q} = 0$  interatomic force constant calculations needed to get the relaxed-atom elastic tensor for metals?

(1) S. de Gironcoli, Phys. Rev. B 51, 6773 (1995)

(2) S. Baroni, S. de Gironcoli, and A. Dal Corso, Rev. Mod. Phys. **73**, 515 (2001)



# Input file for RF run with strain

```
# First dataset : Self-consistent run

# Second dataset : Non-self-consistent run
#                  for full k point set

# Third dataset  : d/dk response calculation
#                  #this section is omitted if
#                  #only the elastic tensor is
#                  #wanted
getwfk3  2
getden3  1
  iscf3  -3
rfelld3  2
  rfdird3 1 1 1

# Fourth dataset : phonon, strain, and homogeneous
#                  electric field response

diemix4  0.85
diemac4  1.0
getwfk4  2
getddk4  3          #omitted for ELT only
  iscf4  3
rfelld4  3          #omitted for ELT only
rfatpol4 1 2
  rfdird4 1 1 1
rfphon4  1
rfstrs4  3          #only this is new for strain

# Common data      #stresses and forces should
  nqpt  1          #(in general) be relaxed
  qpt   0 0 0      #beforehand
```



## 2DTE terms in output file

- Mix of reduced and Cartesian coordinates, also in \_DDB output file
  - With `natom = 2`, electric field `pert = 4` and strain `pert = 5, 6`
  - Only a sample of the complete matrix shown

```
2nd-order matrix (non-cartesian coordinates, masses not included,
asr not included )
cartesian coordinates for strain terms (1/ucvol factor
for elastic tensor components not included)
  j1      j2      matrix element
dir pert dir pert      real part      imaginary part

  1      1      2      2      -2.8200006186      0.0000000000
  1      1      3      2      -2.8654826400      interatomic force constant (red-red)
  1      1      1      4      -4.1367712586      Born effective charge (red-red)
  1      1      2      5      -0.0238530938      internal strain (red-cart)
  1      4      3      4      46.0269881204      dielectric tensor (red-red)
  1      4      3      5      -0.2214090328      piezoelectric tensor (red-cart)
  1      5      2      6      -0.0103809572      elastic tensor (cart-cart)
```

- Cartesian ELT, PZT, and internal strain are also included in the output
- Detailed breakdown of contributions is given for `pertvol = 10`



# Incorporating atomic relaxation

- Implemented as post-processing procedure in anaddb
  - New and modified routines: dielmore9.f, elast9.f, piezo9.f, instr9.f, invars9.f, outvars9.f, diel9.f, anaddb.f, defs\_common.f, defs\_basis.f
- Full theoretical discussion in `Infos/Theory/lr.pdf`
- Basic results:

$$\tilde{C}_{\alpha\beta,\gamma\delta} = C_{\alpha\beta,\gamma\delta} + \Omega^{-1} \sum_{mn=1}^{natom} \sum_{ij=1}^3 \Lambda_{mi,\alpha\beta} (K^{-1})_{mi,nj} \Lambda_{nj,\gamma\delta}$$

$$\tilde{e}_{\alpha\beta,\gamma} = e_{\alpha\beta,\gamma} + \Omega^{-1} \sum_{mn=1}^{natom} \sum_{ij=1}^3 \Lambda_{mi,\alpha\beta} (K^{-1})_{mi,nj} Z_{nj,\gamma}$$

- $\tilde{C}, C$  physical and clamped-atom elastic tensors
- $\tilde{e}, e$  physical and clamped-atom piezoelectric tensors
- $K^{-1}$  pseudo-inverse  $Q=0$  interatomic force constant matrix
- $\Lambda$  internal-strain “force response” tensor
- $Z$  Born effective charge matrix
- All in Cartesian coordinates



# Input file for anaddb run

```
dieflag 3 !flag for relaxed-ion dielectric tensor
elaflag 3 !flag for the elastic tensor
piezoflag 3 !flag for the piezoelectric rensor
instrflag 1 !flag for the internal strain tensor

!the effective charge part
  asr 1
  chneut 1

!Wavevector list number 1

  nph11 1
  qph11 0.0 0.0 0.0 1.0

!Wave vector list no. 2

  nph21 1
  qph21 0.0 0.0 1.0 0.0
```

New flags and/or values in violet

See `Test_v4/t61-70` for more examples



# New output from anaddb

Elastic Tensor(relaxed ion)(Unit:10<sup>2</sup>GP,VOIGT notation):

1.2499151	0.6699976	0.6835944	0.0022847	-0.0113983	-0.0001512
0.6699976	1.6217899	0.5566207	0.0194005	-0.0055653	-0.0055915
0.6835944	0.5566207	1.5896839	-0.0207927	0.0107924	0.0080825
0.0022847	0.0194005	-0.0207927	0.6659339	0.0077398	-0.0056845
-0.0113983	-0.0055653	0.0107924	0.0077398	0.7283916	0.0014049
-0.0001512	-0.0055915	0.0080825	-0.0056845	0.0014049	0.7222881

proper piezoelectric constants(relaxed ion)(Unit:c/m<sup>2</sup>)

0.01714694	0.05107080	-0.00883676
0.00828454	0.03716812	-0.00810176
0.01882065	0.05180658	-0.00576393
-0.03872154	-0.01245206	0.01902693
-0.01424058	0.00757132	-0.00294782
0.01566436	-0.00054740	0.00218470

- Also in output
  - Clamped-ion versions of the above in standard units
  - Clamped and relaxed compliance tensors
  - “Force-response” and “displacement response” internal strain tensors
  - More tensors corresponding to different boundary conditions to be added



# Global comparison with numerical derivatives

- Zinc-blende AIP with random distortions so all tensor elements are non-zero.
  - Ground state calculations of stress and polarization with *exquisitely* relaxed atomic coordinates (but unrelaxed stress)
  - Finite-difference  $d/dk \psi_{\alpha}^{(1)}$ 's for best consistency with polarization calculations
  - Sample of complete set of tensor elements

Elastic Tensor (GPa)

		Numerical	DFPT	Diff
xx	xx	124.991500	124.991500	-1.1E-05
yy	xx	66.999750	66.999760	8.2E-06
zz	xx	68.359440	68.359440	7.0E-07
yz	xx	0.228447	0.228466	1.9E-05
xz	xx	-1.139838	-1.139828	9.6E-06
xy	xx	-0.015028	-0.015117	-9.0E-05
xx	yz	0.228471	0.228466	-4.4E-06
yy	yz	1.940050	1.940054	3.7E-06
zz	yz	-2.079264	-2.079275	-1.1E-05
yz	yz	66.593340	66.593390	5.2E-05
xz	yz	0.773972	0.773977	5.1E-06
xy	yz	-0.568446	-0.568449	-3.2E-06

Piezoelectric Tensor (C/m<sup>2</sup> x 10<sup>-2</sup>)

		Numerical	DFPT	Diff
x	xx	1.714769	1.714694	-7.5E-05
y	xx	5.107069	5.107080	1.1E-05
z	xx	-0.883962	-0.883676	2.9E-04
x	yy	0.828569	0.828454	-1.2E-04
y	yy	3.716843	3.716812	-3.2E-05
z	yy	-0.810201	-0.810176	2.5E-05
x	yz	-3.871980	-3.872154	-1.7E-04
y	yz	-1.245173	-1.245206	-3.3E-05
z	yz	1.902687	1.902693	5.6E-06

- RMS Errors  $4.0 \times 10^{-5}$ , ELT and  $1.7 \times 10^{-6}$ , PZT
  - One-two orders of magnitude smaller errors for clamped-atom quantities.



# Present status, future development

---

- Examples of strain RF and anaddb calculations are `Test_v4/t61-70`
- RF strain is fully parallelized
  - Parallel version was developed simultaneously with sequential
- Present limitations
  - Norm-conserving psp's
  - Non-spin polarized (this is about to be relaxed, testing is nearly complete)
  - LDA only
  - No spin-orbit
- GGA prospects
  - Probably straightforward but complicated by “two kinds of charge” problem with model cores
  - Model core smoothness problem is undoubtedly worse
- Spin-orbit coupling
  - This has all the nonlocal psp complexity, probably significantly worse judging by the existing spin-orbit code for stress
  - *Mathematica* code will eventually be added to the documentation and may help a future developer with this



# Future development, continued

---

- PAW
  - Far beyond norm-conserving psp non-local complexity
  - Needs spherical harmonics with off-diagonal coupling which cannot be turned into dot products with simple metric-tensor dependencies
  - Has “two kinds of charge” problem like model core but much worse, because augmentation charge has non-spherical components

## On the upbeat side, however

- 3<sup>rd</sup>-order response functions involving strain via “2n+1” theorem
  - Require two  $\psi^{(1)}$  and one  $H^{(1)}$ , all available
  - Eg., electrostriction, non-linear elastic constants, Grüneisen parameters
- It’s time for feedback – let’s see what the users want and what trouble they get into
  - If a user wants a strain feature badly enough *we’ll have a new developer!*
  - Isn’t that the ABINIT philosophy?



# Appendix : *Mathematica* for nonlocal psp

$$\langle \mathbf{K}' | V_{NL} | \mathbf{K} \rangle = \frac{1}{\Omega} \sum_{\kappa \ell m} e^{i\mathbf{K}' \cdot \boldsymbol{\tau}_\kappa} f_{\kappa \ell}(\mathbf{K}' \cdot \mathbf{K}') \mathcal{P}_\ell(\mathbf{K}' \cdot \mathbf{K}', \mathbf{K}' \cdot \mathbf{K}, \mathbf{K} \cdot \mathbf{K}) e^{-i\mathbf{K} \cdot \boldsymbol{\tau}_\kappa} f_{\kappa \ell}(\mathbf{K} \cdot \mathbf{K}).$$

- Define tensor products  $T_{\ell m}(\tilde{\mathbf{K}})$  and  $T_{\ell m}(\tilde{\mathbf{K}}')$ 
  - Follows D. C. Allan

```
tnk = {
{1}, (* 0, 1 *)
{k1, (* 1, 1 *)
k2, (* 1, 2 *)
k3}, (* 1, 3 *)

{k1 k1, (* 2, 1 *)
k2 k2, (* 2, 2 *)
k3 k3, (* 2, 3 *)
k3 k2, (* 2, 4 *)

etc. to rank 7
```

- Define  $K$ 's, metric tensor functions, dot products, and Legendre's
- $s_1$  and  $s_2$  are strain variables

```
k = {k1, k2, k3}; kp = {kp1, kp2, kp3};
m = {{m11[s1,s2], m12[s1,s2], m13[s1,s2]},
{m12[s1,s2], m22[s1,s2], m23[s1,s2]},
{m13[s1,s2], m23[s1,s2], m33[s1,s2]}};
dt = kp.m.k; ks = k.m.k; kps = kp.m.kp;
Plegendre={1, dt, 1.5 dt^2 - 0.5 kps ks,
2.5 dt^3 - 1.5 kps dt ks};
```



# Mathematica for nonlocal psp, continued

- Strain derivatives of form factors  $f_\ell$  “bring out” derivatives of dot products
- Define 6 combinations of dot product derivatives and Legendre derivatives that have given offsets between “input” and “output” rank

<pre>poly = {D[kps,s2] D[kps,s1] Plegendre[[rank+1]],         D[ks, s2] D[ks, s1] Plegendre[[rank+1]],         (D[D[kps,s2],s1] Plegendre[[rank+1]] + D[kps,s1] D[Plegendre[[rank+1]],s2] + D[kps,s2] D[Plegendre[[rank+1]],s1]),         (D[D[ks, s2],s1] Plegendre[[rank+1]] + D[ks, s1] D[Plegendre[[rank+1]],s2] + D[ks, s2] D[Plegendre[[rank+1]],s1]),         (D[kps,s1] D[ks,s2] + D[kps,s2] D[ks,s1])         Plegendre[[rank+1]],         D[D[Plegendre[[rank+1]],s2],s1]};</pre>	$l \rightarrow l+4$ $l+4 \rightarrow l$	$f_\ell, f_\ell''$ $f_\ell'', f_\ell$
	$l \rightarrow l+2$	$f_\ell, f_\ell'$
	$l+2 \rightarrow l$	$f_\ell', f_\ell$
	$l+2 \rightarrow l+2$ $l \rightarrow l$	$f_\ell', f_\ell'$ $f_\ell, f_\ell$

– In *Mathematica*  $df/dx$  is  $D[f, x]$

- Now, do the work – extract the coefficients of each pair of input and output tensors

```
Do[term = Simplify[Coefficient[poly[[item]], (tnkp[[rankout+1]][[jj]] *
tnk[[rankin+1]][[ii]])];
```



# Title

---

- X

- X

- X

- X

