# Adaptively Compressed Polarizability Operator

## For Accelerating Large Scale *ab initio* Phonon Calculations

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# What is Phonon?

*"In physics, a phonon is a collective excitation in a periodic, elastic arrangement of atoms or molecules in condensed matter... it represents an excited state in the quantum mechanical quantization of the modes of vibrations of elastic structures of interacting particles."*

—wikipedia

# Lattice Wave

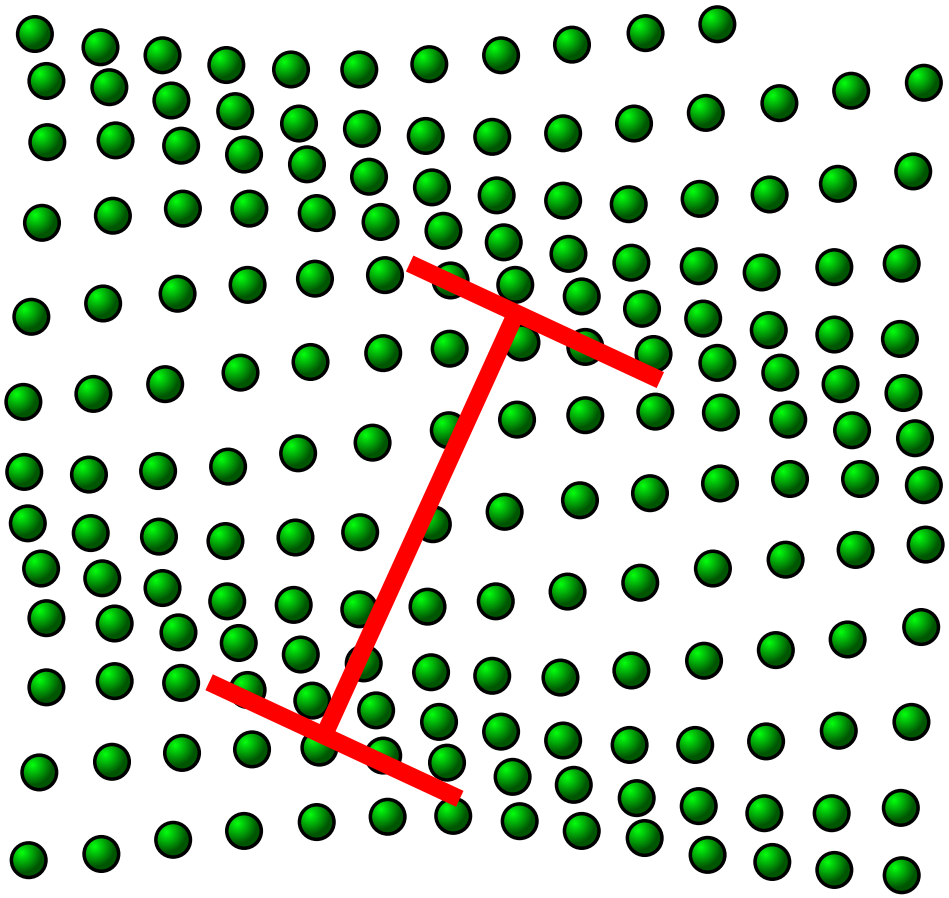

Figure 1: Lattice wave example. Figure from wikipedia.

# Phonon Dispersion Relations

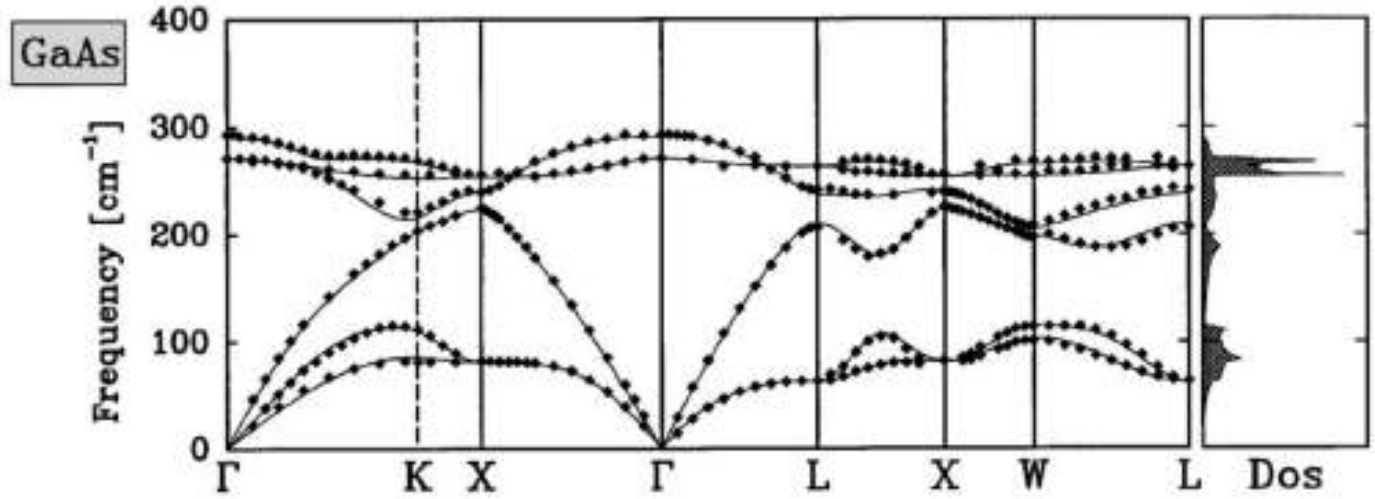


Figure 2: Phonon dispersion and densities of states of gallium arsenide (GaAs). Figure from S. Baroni *et al.*, 2001 .

# Why Care?

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- Thermal conductivity; specific heat;

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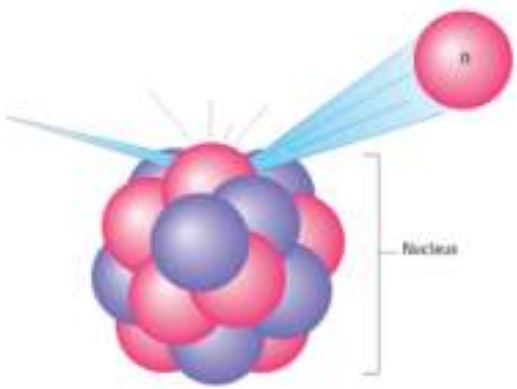


Figure 3: In elastic neutron-scattering, the neutron bounces off the bombarded nucleus without exciting or destabilizing it. Figure from The Schlumberger Oilfield Glossary.

# Electron-phonon interaction

- Thermal conductivity; specific heat;
- Elastic neutron scattering;
- Electrical conductivity;
- Electron-phonon interaction related topics;
  - major contributor to electrical resistance in most inorganic metals and semiconductors above zero (very low) temperature;
  - Overheating problem;

# Dynamical Matrix

$$D_{I,J} = \frac{1}{\sqrt{M_I M_J}} \frac{\partial^2 E_{\text{tot}}(\{\mathbf{R}_I\})}{\partial \mathbf{R}_I \partial \mathbf{R}_J},$$

- $M_I$ mass of the $I$-th atom, $I = 1, \ldots, N_A$.
- $\mathbf{R}_I \in \mathbb{R}^d$ position of the $I$-th atom, $I = 1, \ldots, N_A$.
- $D \in \mathbb{R}^{dN_A \times dN_A}$

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# Density Functional Theory

*Ab initio* calculation – DFT.

- most widely used;
- exact description of ground state properties:
- electron density, energy, and atomic forces.

# Kohn-Sham Density Functional Theory

$$
\begin{aligned}
& E_{\mathrm{KS}}(\{\psi_i\};\{\mathbf{R}_I\}) \\
=& \frac{1}{2}\sum_{i=1}^{N_e}\int |\nabla\psi_i(\mathbf{r})|^2\,\mathrm{d}\mathbf{r} + \int V_{\mathsf{ion}}(\mathbf{r};\{\mathbf{R}_I\})\rho(\mathbf{r})\,\mathrm{d}\mathbf{r} \\
& + \frac{1}{2}\iint v_c(\mathbf{r},\mathbf{r}')\rho(\mathbf{r})\rho(\mathbf{r}')\,\mathrm{d}\mathbf{r}\,\mathrm{d}\mathbf{r}' + E_{\mathrm{xc}}[\rho] \\
& + E_{\mathrm{II}}(\{\mathbf{R}_I\}).
\end{aligned}
\tag{1}
$$

# Derivative

$$\mathbf{F}_I = -\int \frac{\partial V_I}{\partial \mathbf{R}_I}(\mathbf{r}-\mathbf{R}_I)\rho(\mathbf{r})\,\mathrm{d}\mathbf{r} - \frac{\partial E_{\mathrm{II}}(\{\mathbf{R}_I\})}{\partial \mathbf{R}_I}.$$

$$\frac{\partial^2 E_{\mathsf{tot}}(\{\mathbf{R}_I\})}{\partial \mathbf{R_I}\partial \mathbf{R_J}}$$
$$= \int \frac{\partial V_I}{\partial \mathbf{R}_I}(\mathbf{r}-\mathbf{R}_I)\frac{\delta\rho(\mathbf{r})}{\delta \mathbf{R}_J}\,\mathrm{d}\mathbf{r} + \delta_{I,J}\int \rho(\mathbf{r})\frac{\partial^2 V_I}{\partial \mathbf{R}_I^2}(\mathbf{r}-\mathbf{R}_I)\,\mathrm{d}\mathbf{r}$$
$$+ \frac{\partial^2 E_{\mathrm{II}}(\{\mathbf{R}_I\})}{\partial \mathbf{R}_I \partial \mathbf{R}_J}.$$

# Derivative

$$\mathbf{F}_I = -\int \frac{\partial V_I}{\partial \mathbf{R}_I}(\mathbf{r}-\mathbf{R}_I)\rho(\mathbf{r})\,\mathrm{d}\mathbf{r} - \frac{\partial E_{\mathrm{II}}(\{\mathbf{R}_I\})}{\partial \mathbf{R}_I}. \tag{2}$$

$$\begin{aligned}&\frac{\partial^2 E_{\mathsf{tot}}(\{\mathbf{R}_I\})}{\partial \mathbf{R_I}\partial \mathbf{R_J}}\\ =&\int \frac{\partial V_I}{\partial \mathbf{R}_I}(\mathbf{r}-\mathbf{R}_I)\frac{\delta\rho(\mathbf{r})}{\delta \mathbf{R}_J}\,\mathrm{d}\mathbf{r} + \delta_{I,J}\int \rho(\mathbf{r})\frac{\partial^2 V_I}{\partial \mathbf{R}_I^2}(\mathbf{r}-\mathbf{R}_I)\,\mathrm{d}\mathbf{r}\\ &+\frac{\partial^2 E_{\mathrm{II}}(\{\mathbf{R}_I\})}{\partial \mathbf{R}_I\partial \mathbf{R}_J}.\end{aligned} \tag{3}$$

# Polarizability Operator

$$\frac{\delta\rho(\mathbf{r})}{\delta\mathbf{R}_J} = \int \frac{\delta\rho(\mathbf{r})}{\delta V_{\text{ion}}(\mathbf{r}')} \frac{\partial V_J}{\partial \mathbf{R}_J}(\mathbf{r}' - \mathbf{R}_J)\,\mathrm{d}\mathbf{r}'.$$

Fréchet derivative $\chi(\mathbf{r}, \mathbf{r}') := \dfrac{\delta\rho(\mathbf{r})}{\delta V_{\text{ion}}(\mathbf{r}')}$ is the reducible polarizability operator.

# KSDFT Cont.

Kohn-Sham equations (W. Kohn and L. Sham 1965)

$$H[\rho]\psi_i = \left(-\frac{1}{2}\Delta + \mathcal{V}[\rho]\right)\psi_i = \varepsilon_i\psi_i, \tag{4}$$

$$\int \psi_i(\mathbf{r})\psi_j(\mathbf{r})\,\mathrm{d}\mathbf{r} = \delta_{ij}, \quad \rho(\mathbf{r}) = \sum_{i=1}^{N_e} |\psi_i(\mathbf{r})|^2 . \tag{5}$$

- $\varepsilon_i$ energy level; $\psi_i$ orbitals;
- index $i = 1, \ldots, N_e$ called occupied states; $i = N_e + 1, \ldots$ unoccupied states;
- $\varepsilon_g = \varepsilon_{N_e+1} - \varepsilon_{N_e}$ band gap;
- $\mathcal{V}[\rho](\mathbf{r}) = V_{\text{ion}}(\mathbf{r}; \{\mathbf{R}_I\}) + \int v_c(\mathbf{r}, \mathbf{r}')\rho(\mathbf{r}')\,\mathrm{d}\mathbf{r}' + V_{\text{xc}}[\rho](\mathbf{r})$
- $V_{\text{ion}} = \sum_I V_I, \quad g_J = \frac{\partial V_J}{\partial \mathbf{R}_J}$.

# Sternheimer Equations

$$
\begin{aligned}
&Q(\varepsilon_i - H)Q\zeta_{ij} = Q(\psi_i \odot g_j). \\
&i = 1, \ldots, N_e, j = 1, \ldots, dN_a
\end{aligned}
\tag{6}
$$

- $Q$ projection onto unoccupied states;
- $\zeta_{ij}$ defined as the solution to the equation.

This is the core part of density-functional perturbation theory (DFPT) (S. Baroni *et al.* 2001).

# How to solve that?

$$Q(\varepsilon_i - H)Q\zeta_{ij} = Q(\psi_i \odot g_j).$$
$$i = 1, \ldots, N_e, j = 1, \ldots, dN_a$$

- SVD?
- Frozen phonon approach? (Or finite difference in math)
- Compression?

# How to solve that? cont.

$$Q(\varepsilon_i - H)Q\zeta_{ij} = Q(\psi_i \odot g_j).$$
$$i = 1, \ldots, N_e, j = 1, \ldots, dN_a$$

- SVD?
- Frozen phonon approach? (Or finite difference in math)
- Compression!

ACP formulation reduces the computational complexity of phonon calculations from $\mathcal{O}(N_e^4)$ to $\mathcal{O}(N_e^3)$ for the first time (**X**.*et al.* 2016).

# The Dyson Equation

Irreducible polarizability operator $\chi_0 = \frac{\delta \rho}{\delta \mathcal{V}}$

$$\begin{aligned} \chi &= \chi_0 + \chi_0 v_{\text{hxc}} \chi \\ \text{or} \quad U &= \chi_0 G + \chi_0 v_{\text{hxc}} U, \quad U = \chi G. \end{aligned} \tag{7}$$

So compression of $\chi_0$ can only be done *adaptively*.

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# Adaptively Compressed Polarizability Operator

$$Q(\varepsilon_i - H)Q\zeta_{ij} = Q(\psi_i \odot g_j).$$
$$i = 1, \ldots, N_e, j = 1, \ldots, dN_a$$

- Chebyshev interpolation – disentangle the energy dependence on the left
- Interpolative separable density fitting method – compress the rhs vectors.
- Adaptively compressed – cost stays the same across iterations.

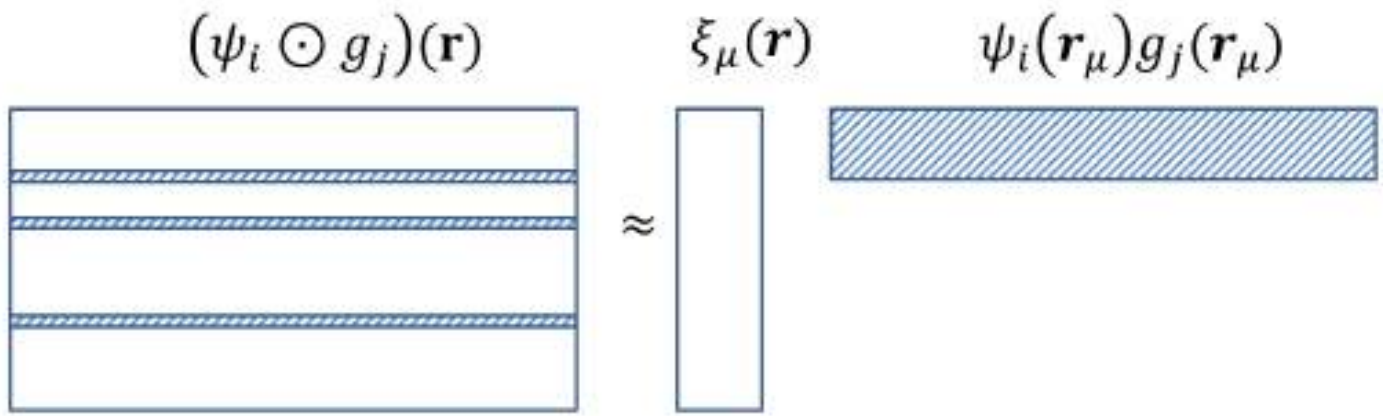
# Chebyshev Interpolation

Pick $\{\varepsilon_c\} \in \mathcal{I} \equiv [\varepsilon_1, \varepsilon_{N_e}]$.
Lagrange interpolation

$$\zeta = \sum_{c=1}^{N_c} \widetilde{\zeta}_c \prod_{c' \neq c} \frac{\varepsilon - \widetilde{\varepsilon}_{c'}}{\widetilde{\varepsilon}_c - \widetilde{\varepsilon}_{c'}}.$$

# Interpolative Decomposition

$$M_{ij} = \psi_i \odot g_j, \quad \text{or} \quad M_{ij}(\mathbf{r}) = \psi_i(\mathbf{r}) g_j(\mathbf{r}).$$

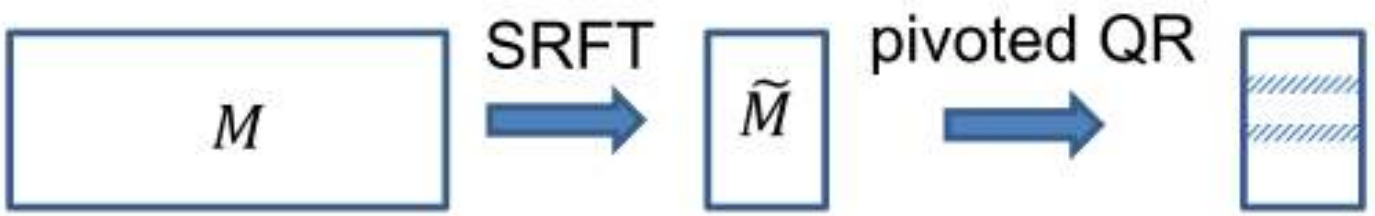



$$M_{ij}(\mathbf{r}) \approx \sum_{\mu=1}^{N_\mu} \xi_\mu(\mathbf{r}) M_{ij}(\mathbf{r}_\mu) \equiv \sum_{\mu=1}^{N_\mu} \xi_\mu(\mathbf{r}) \psi_i(\mathbf{r}_\mu) g_j(\mathbf{r}_\mu). \tag{8}$$

(H. Cheng, Z. Gimbutas, P. G. Martinsson, and V. Rokhlin, 2005)

# Interpolative Separable Density Fitting

Two-step procedure: subsampled random Fourier Transform and QR decomposition.




(J. Lu and L. Ying, 2015)

# Reconstruction

To avoid $\mathcal{O}(N_e^4)$ complexity, construct

$$W_\mu = 2\sum_{i=1}^{N_e} \psi_i \odot \left( \sum_{c=1}^{N_c} \widetilde{\zeta}_{c\mu} \prod_{c' \neq c} \frac{\varepsilon_i - \widetilde{\varepsilon}_{c'}}{\widetilde{\varepsilon}_c - \widetilde{\varepsilon}_{c'}} \right) \psi_i(\mathbf{r}_\mu). \tag{9}$$

$$\chi_0 g_j \approx \sum_{\mu=1}^{N_\mu} W_\mu g_j(\mathbf{r}_\mu), \tag{10}$$

or formally, we have $\chi_0 \approx \widetilde{\chi_0} := W\Pi^T$.

# Iterative scheme

Introduce the following change of variable

$$U = \widetilde{U} - B, \quad B = v_{\mathrm{hxc}}^{-1} G, \tag{11}$$

$$\widetilde{U} = \widetilde{\chi}_0[\widetilde{U}] v_{\mathsf{hxc}} \widetilde{U} + B. \tag{12}$$

Iterative scheme:

$$\begin{aligned} &(a)\text{Construct} \quad \chi_0^k[\widetilde{U}^k] = W^k (\Pi^k)^T \\ &(b)\widetilde{U}^{k+1} = \left(I - W^k (\Pi^k)^T v_{\mathsf{hxc}}\right)^{-1} B \\ &\qquad = B + W^k \left(I - (\Pi^k)^T v_{\mathsf{hxc}} W^k\right)^{-1} (\Pi^k)^T v_{\mathsf{hxc}} B. \end{aligned} \tag{13}$$

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# One-dimensional Model

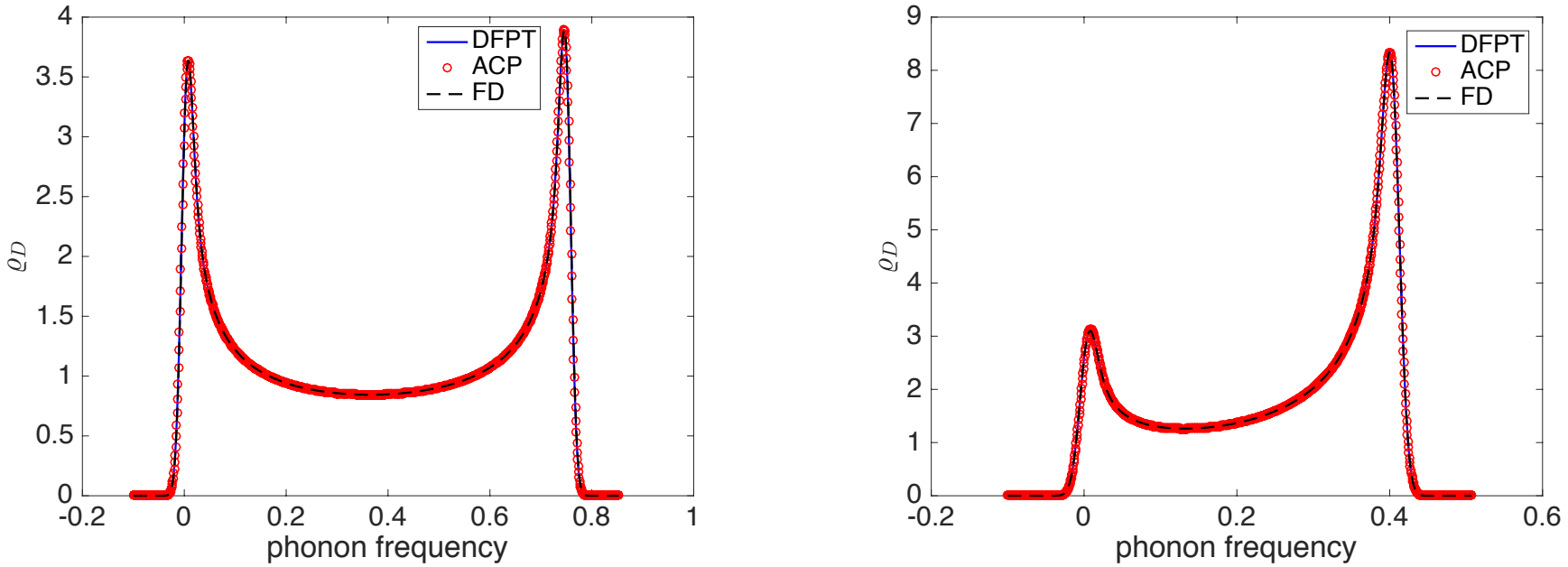



Figure 4: Phonon spectrum for the 1D systems computed using ACP, DFPT, and FD, for both (a) insulating and (b) semiconducting systems.

# One-dimensional Model cont.

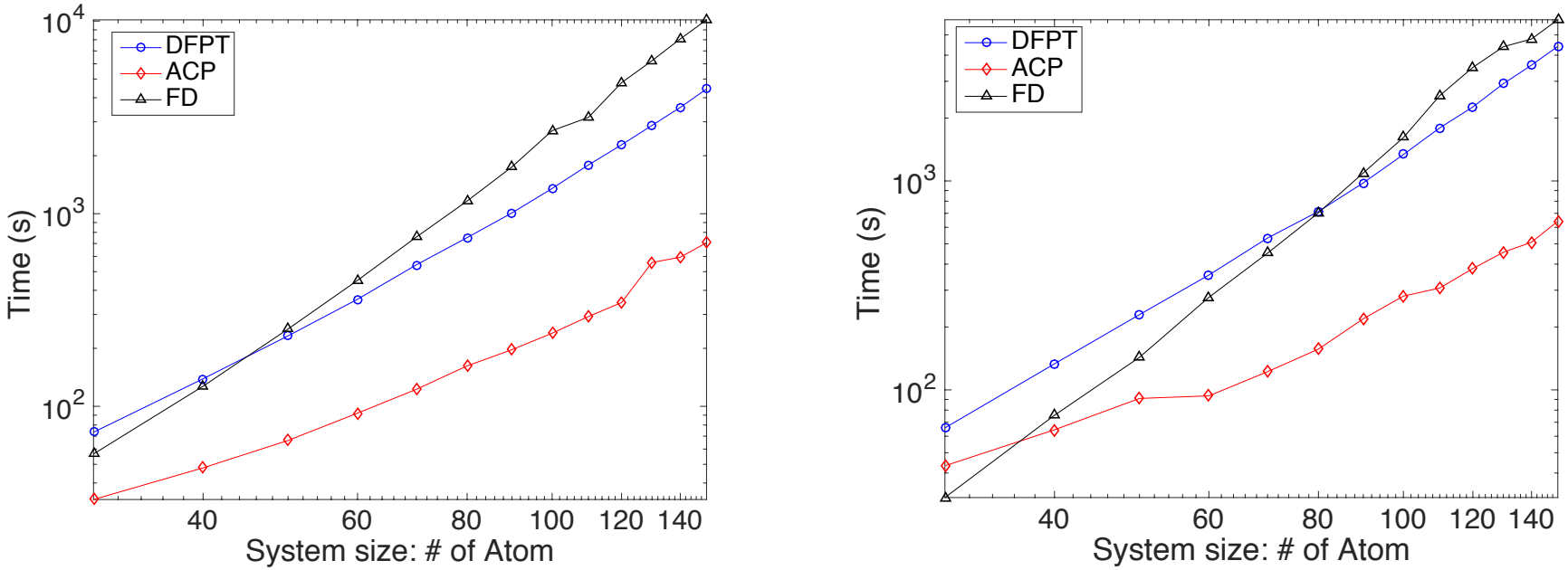
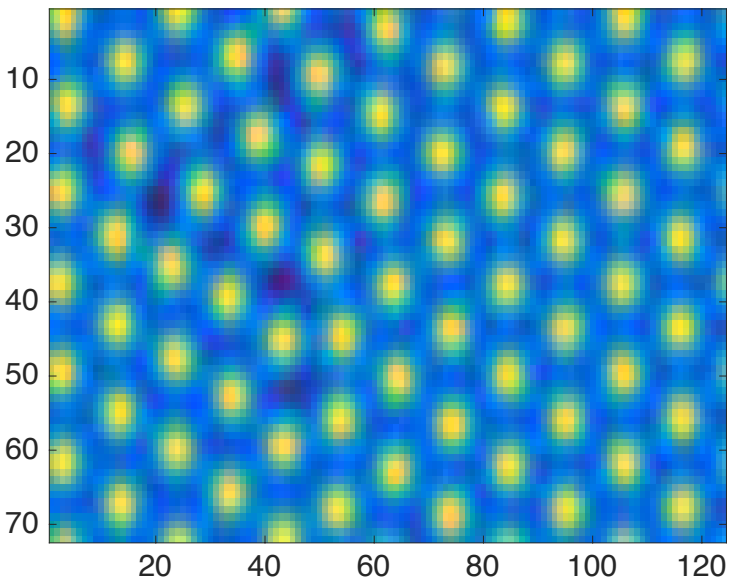



Figure 5: Computational time of 1D examples. Comparison among DFPT, ACP, and FD for (a) insulating, and (b) semiconducting systems, respectively.

# Two-dimensional Model



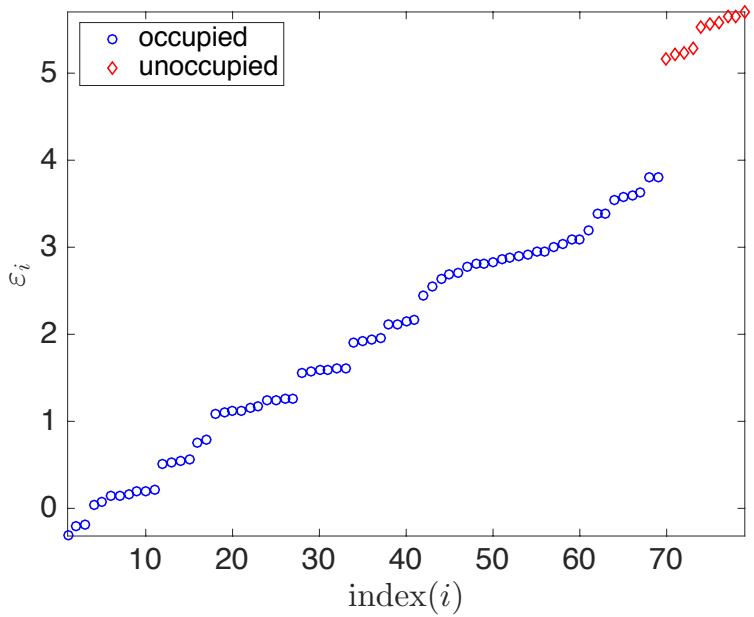




Figure 6: The electron density $\rho$ of the 2D system with defects (a), and the occupied and unoccupied eigenvalues (b).

# Two-dimensional Model cont.

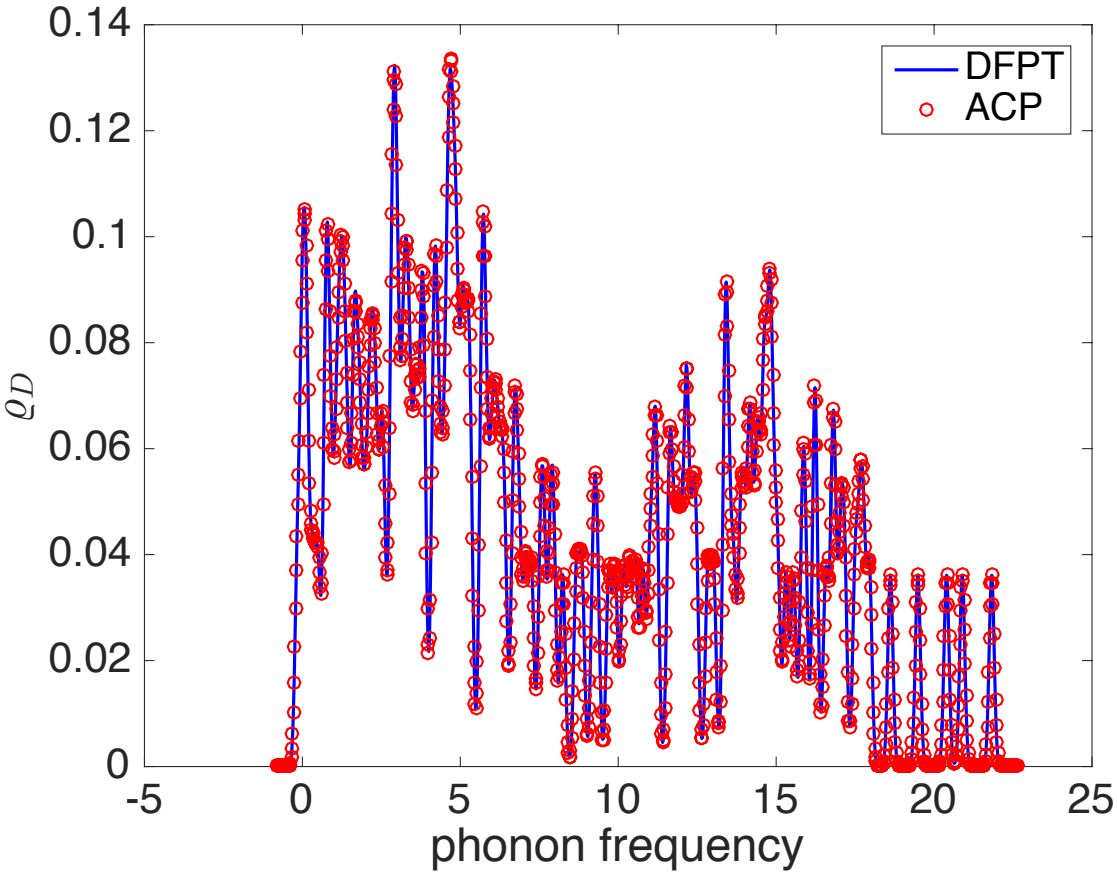



Figure 7: Phonon spectrum for the 2D system with defects. $N_A = 69$.

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# Split Representation of ACP

$$\chi_0(\mathbf{r},\mathbf{r}') = 2\sum_{i=1}^{N_e}\sum_{j=N_e+1}^{\infty}\frac{\psi_i(\mathbf{r})\psi_j(\mathbf{r})\psi_i(\mathbf{r}')\psi_j(\mathbf{r}')}{\varepsilon_i-\varepsilon_j}$$

$$= 2\left[\sum_{i=1}^{N_e}\sum_{j=N_e+1}^{N_t}\frac{\psi_i(\mathbf{r})\psi_j(\mathbf{r})\psi_i(\mathbf{r}')\psi_j(\mathbf{r}')}{\varepsilon_i-\varepsilon_j} + \sum_{i=1}^{N_e}\sum_{j=N_t+1}^{\infty}\frac{\psi_i(\mathbf{r})\psi_j(\mathbf{r})\psi_i(\mathbf{r}')\psi_j(\mathbf{r}')}{\varepsilon_i-\varepsilon_j}\right]$$

$$:= \chi_0^{(1)} + \chi_0^{(2)}.$$

# Silicon with 8 Atoms

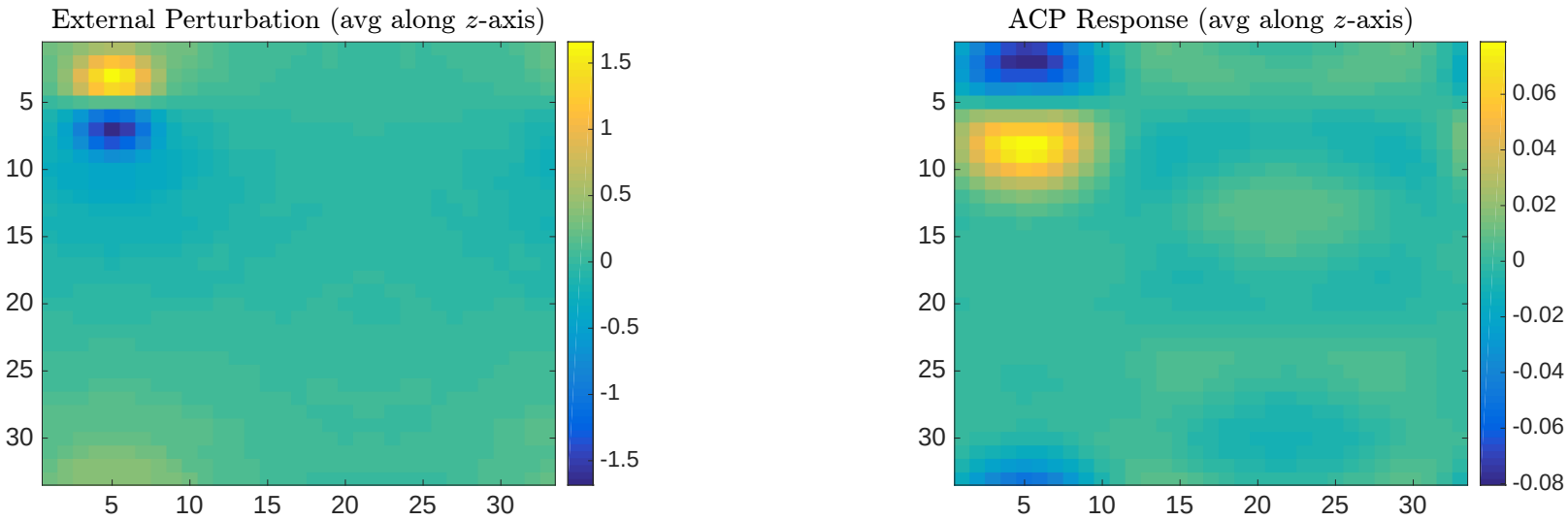
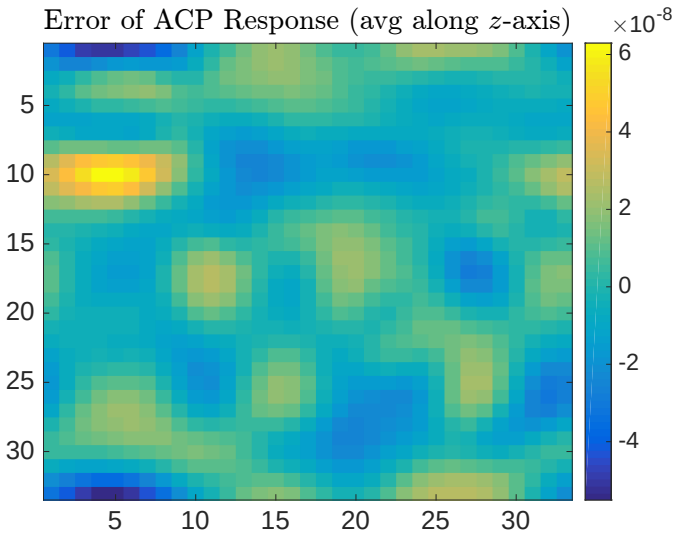



Figure 8: External perturbation and response.

# Silicon with 8 Atoms cont.



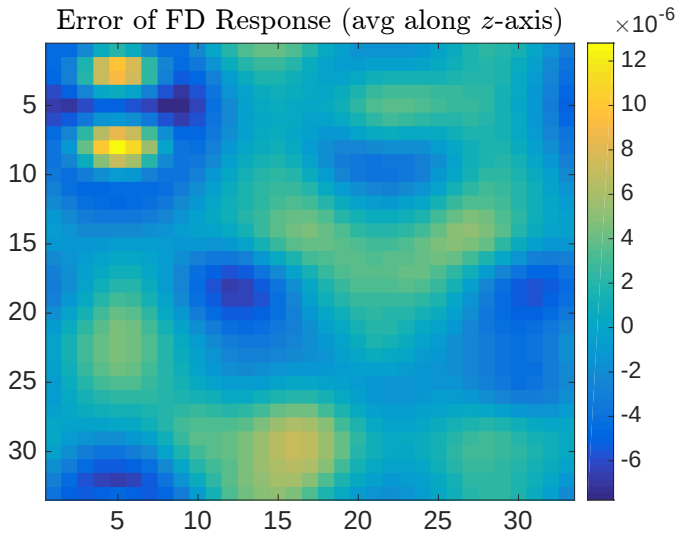




Figure 9: Error on response.

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# Conclusion

- ACP formulation reduces the computational complexity of phonon calculations from $\mathcal{O}(N_e^4)$ to $\mathcal{O}(N_e^3)$ for the first time.
- Recently we extend the formulation to cope with real materials.

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Thanks for your attention.