Implementation and analysis of a plane wave and real space pseudopotential method including an efficient spin-orbit coupling treatment tailored to calculate the electronic structure of large-scale semiconductor nanostructures

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Applications

- Optoelectronic devices
- Quantum information

Nanostructures

- Quantum wells, wires & dots
- Nano, but ... 10^3 – 10^5 atoms

Capabilities of current atomistic methods (DFT): 1000 – 5000 atoms at most

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Screened effective pseudopotentials

- No self-consistency cycle as in DFT
- No total energy

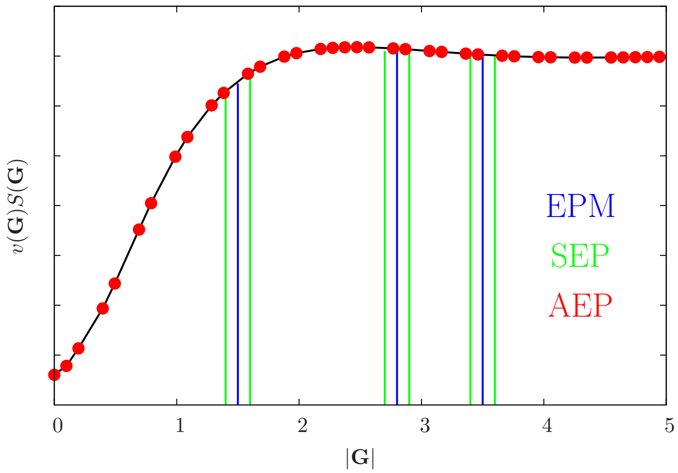
Empirical pseudopotential method

$$V(\boldsymbol{r}) = \sum_{\boldsymbol{G}} v(\boldsymbol{G}) S(\boldsymbol{G}) e^{i\boldsymbol{G}\boldsymbol{r}}$$

Semi-empirical pseudopotentials

DFT, different structures & lattice constants

Atomic Effective Pseudopotentials



Large-scale calculations

- Few selected eigenstates of eigenvalue spectrum
  Electronic & optical properties ↔ States close to VB & CB
- Real space implementation → $\mathcal{O}(N)$

▸ Large-scale Atomic Effective Pseudopotential Program (LATEPP)

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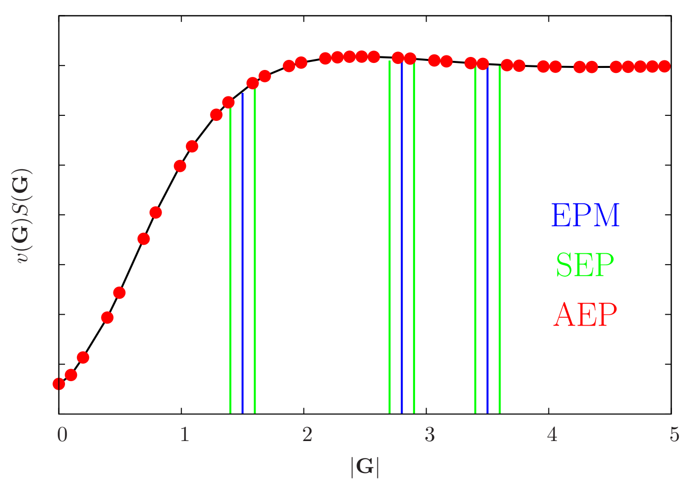


Atomic Effective Pseudopotential — AEP

Basic idea

Screened local effective crystal potential from self-consistent DFT

$$V_{\text{eff}}^{\text{sc}} = V_{\text{local}}^{\text{Pseudo}} + V_{\text{Hartree}} + V_{\text{xc}}$$

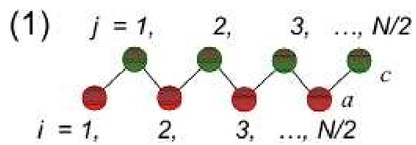
derive

Atomic Effective Pseudopotentials

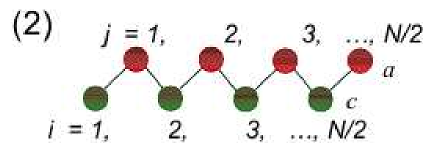
$$V_{\text{eff}}^{\text{sc}}(\boldsymbol{r}) = \sum_{\alpha,n} v_\alpha(\boldsymbol{r} - \boldsymbol{\tau}_{\alpha,n}) \approx \sum_{\alpha,n} v_\alpha(|\boldsymbol{r} - \boldsymbol{\tau}_{\alpha,n}|)$$

Formalism for extended supercells

Anion & cation potentials: $v_\pm = v_a \pm v_c$



$$V_{\text{eff}}^{(1)}(\mathbf{r}) = \sum_{i=1}^{N/2} v_a(\mathbf{r}_i) + \sum_{j=1}^{N/2} v_c(\mathbf{r}_j)$$



$$V_{\text{eff}}^{(2)}(\mathbf{r}) = \sum_{j=1}^{N/2} v_a(\mathbf{r}_j) + \sum_{i=1}^{N/2} v_c(\mathbf{r}_i)$$

Analytic connection by Fourier transform

$$F[V_{\text{eff}}^{(\pm)}(\mathbf{r})] = \frac{1}{\Omega}\int_\Omega [V_{\text{eff}}^{(1)}(\mathbf{r}) \pm V_{\text{eff}}^{(2)}(\mathbf{r})] e^{-i\mathbf{G}\cdot\mathbf{r}} d\mathbf{r} = \beta_\pm v_\pm(\mathbf{G})$$

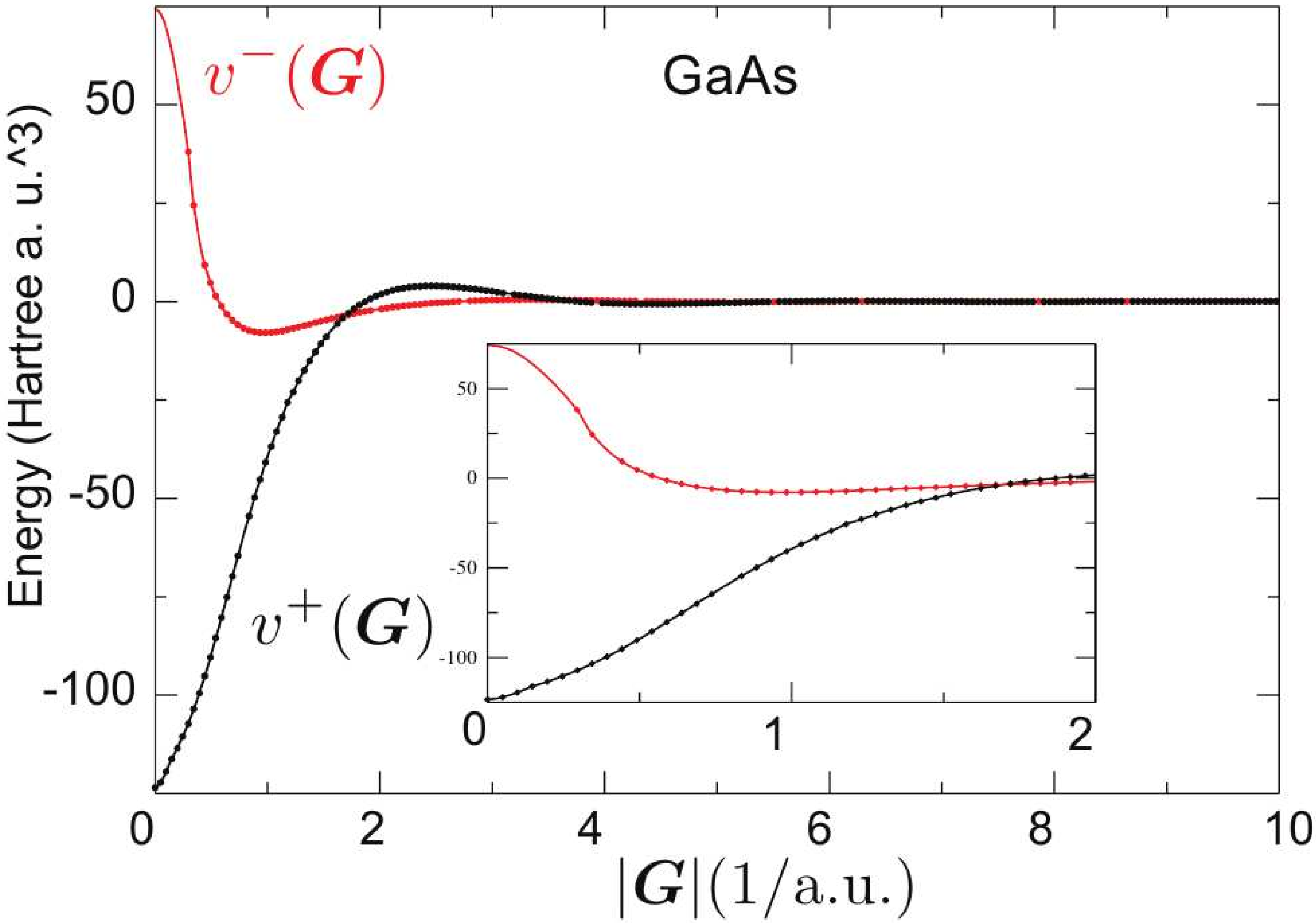
$$V_{\text{eff}}^{(1)}(\mathbf{r}) \pm V_{\text{eff}}^{(2)}(\mathbf{r}) = \sum_{n=1}^{N} (\pm 1)^{n+1} v_\pm(\mathbf{r}_n)$$

Spherical approximation

$$v_\pm(|\mathbf{G}|) = \Re[v_\pm(\mathbf{G})] = f(\beta_\pm, F[V_{\text{eff}}^{(\pm)}(\mathbf{r})])$$

J. R. Cárdenas and G. Bester
Phys. Rev. B **86** 115332 (2012)

From elongated & slightly deformed supercells



AEPs applied to heterostructures show

- nice agreement with self-consistent DFT results
- excellent transferability

√

$V_{\text{eff}}^{(1)}(\mathbf{r}) \pm V_{\text{eff}}^{(2)}(\mathbf{r}) = \sum_{n=1} (\pm 1)^{n+}$

Spherical approximation

$v_\pm(|\mathbf{G}|) = \Re[v_\pm(\mathbf{G})] = f(\beta_\pm, F[$

Large-Scale Atomic Effective Pseudopotential Program

$$\left(-\frac{\nabla^2}{2} + V_{\mathrm{L}} + \widehat{\mathrm{V}}_{\mathrm{NL}} + \widehat{\mathrm{V}}_{\mathrm{SO}}\right)\Psi_{i,\boldsymbol{k}} = \epsilon_{i,\boldsymbol{k}}\Psi_{i,\boldsymbol{k}}$$

Kinetic energy

- $T_{\boldsymbol{G},\boldsymbol{G}'}(\boldsymbol{k}) = \langle \boldsymbol{k}+\boldsymbol{G}|T|\boldsymbol{k}+\boldsymbol{G}'\rangle = \delta_{\boldsymbol{G},\boldsymbol{G}'}\frac{1}{2}(\boldsymbol{k}+\boldsymbol{G})^2$
- Real space: Finite difference scheme $\rightarrow \mathcal{O}(N)$

Local effective potential V_{L}

- $V^{\mathrm{loc,eff}}(\boldsymbol{G}) = \frac{1}{\Omega_{\mathrm{c}}}\sum_{\alpha}^{N_{\mathrm{species}}} v_\alpha(\boldsymbol{G})\sum_{n}^{N_\alpha}\exp\left(-i\boldsymbol{G}\boldsymbol{\tau}_{\alpha,n}\right)w_{\alpha,n}$
- Iterative solvers $\xrightarrow{\mathrm{FFT}} V^{\mathrm{loc,eff}}(\boldsymbol{r})\Psi_{i,\boldsymbol{k}}(\boldsymbol{r})$

Nonlocal potential $\widehat{\mathrm{V}}_{\mathrm{NL}}$

- Fully separable formulation of Kleinman & Bylander
$$\sum_{l,m}|l,m\rangle\delta V_l(\boldsymbol{r})\langle l,m| \xrightarrow{\mathrm{KB}} \sum_{l,m}|\chi_{lm}^{\mathrm{KB}}\rangle E_l^{\mathrm{KB}}\langle\chi_{lm}^{\mathrm{KB}}|$$
- Evaluation in real or reciprocal space

Spin-Orbit potential $\widehat{\mathrm{V}}_{\mathrm{SO}}$

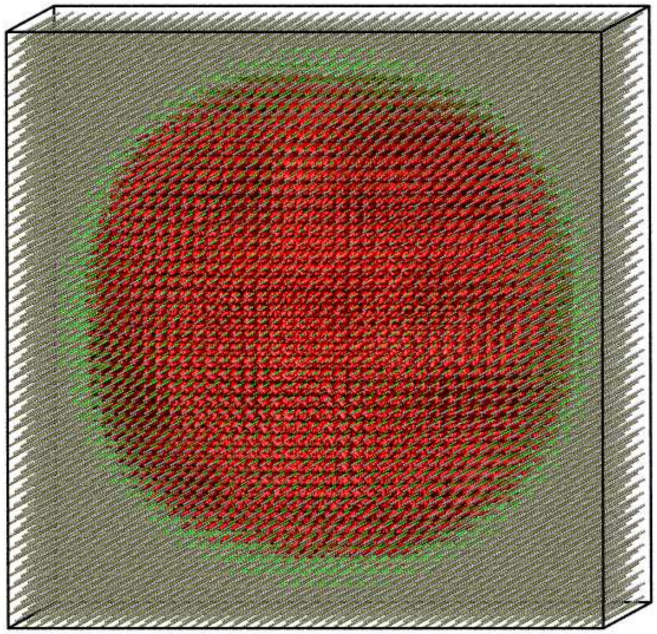
- Spin-Orbit coupling in norm-conserving pseudopotential
$$\widehat{\mathrm{V}}_{\mathrm{SO}} = \sum_{l,m}|l,m\rangle\left[V_l^{\mathrm{SO}}(\boldsymbol{r})\boldsymbol{L}\cdot\boldsymbol{S}\right]\langle l,m|$$
- Scalar relativistic part included in pseudopotential
- Efficient real space evaluation in spin angular function basis: $|l,m,s=\pm 1/2\rangle \rightarrow |j=l\pm 1/2, M\rangle$

LATEPP

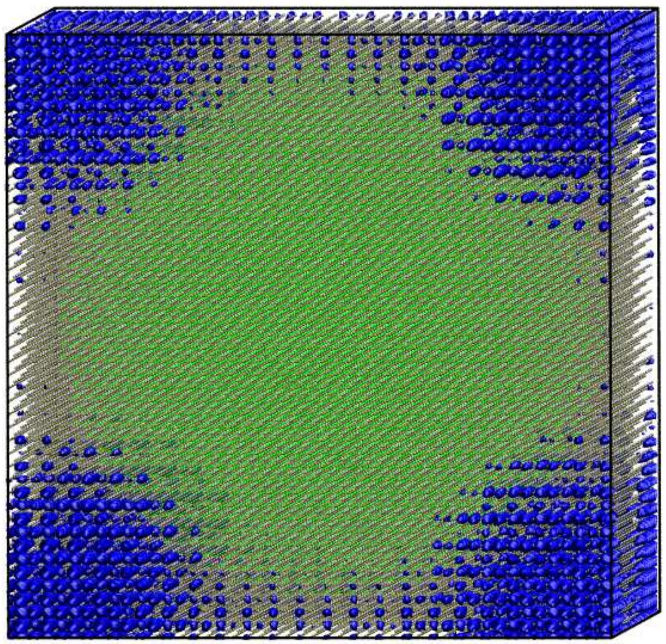
- $\Psi_{i,\boldsymbol{k}}$ represented & evaluated
  - in plane wave basis
  - on real space grid
- Reciprocal space $\leftrightarrow$ Real space
  FFTW & FFTE
- Weights $w_{\alpha,n}$
  for alloys & interfaces
- Atomic Effective Potentials
  No self-consistency required
- Compute few eigenstates
  - Iterative solver
  - Arnoldi restart method (ARPACK)
  - Eigenstates around E_{ref}
    $H\Psi \rightarrow H\Psi - E_{\mathrm{ref}}\Psi$

  $\Rightarrow$ Large-scale eigenproblems
- OpenMP parallelization

Large-Scale Atomic Effective Pseudopotential Program



Highest occupied states

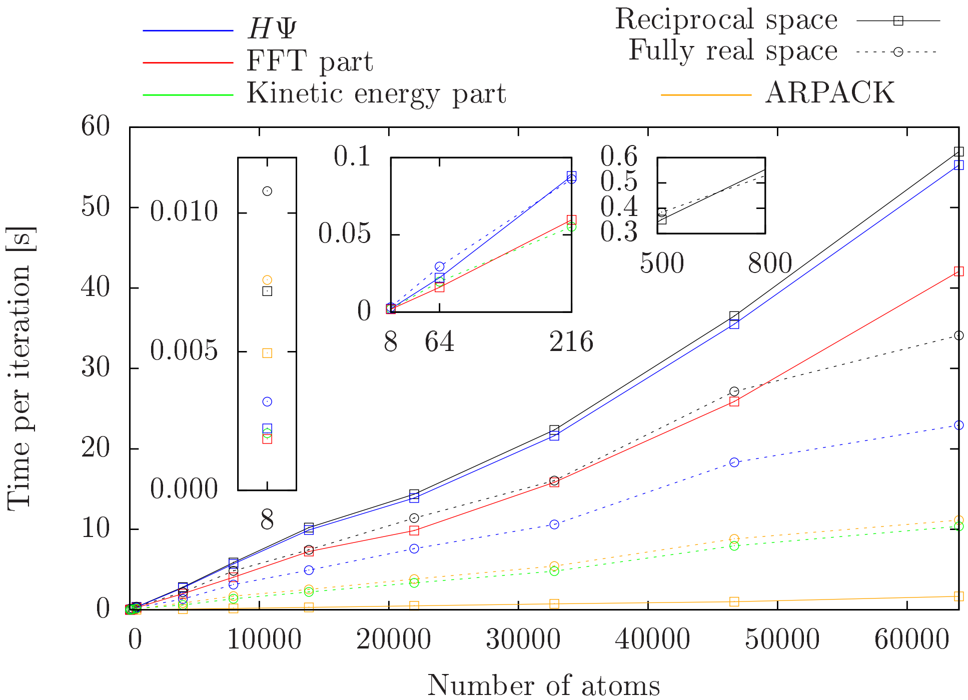


Lowest unoccupied states

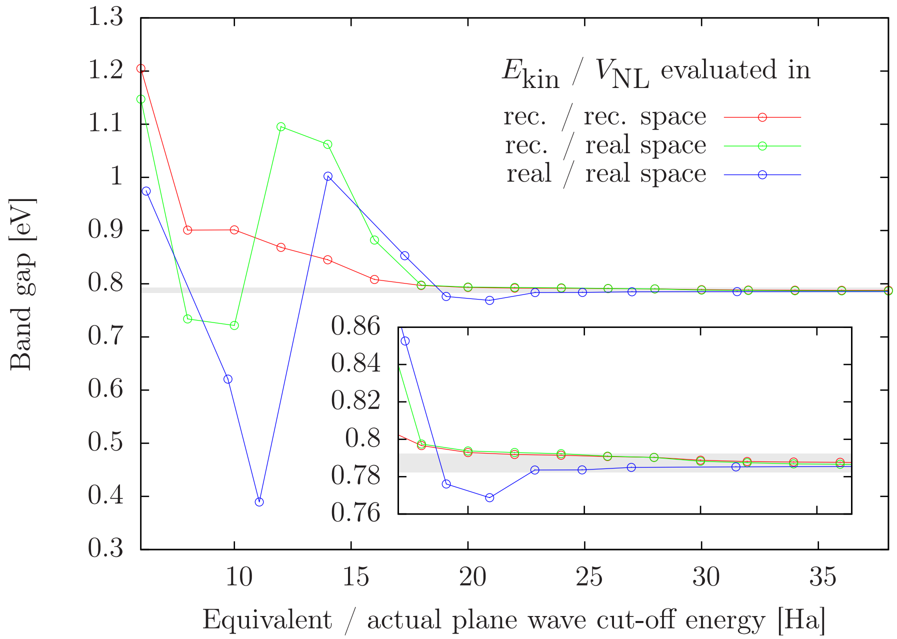
GaAs quantum dot in AlAs

- GaAs QD radius: 6 nm
- AlAs host: 23 lattice constants
- Total: 97,336 atoms
- 31 million plane waves
- 20 states close to CB & VB
- Single node

Scaling



Large-Scale Atomic Effective Pseudopotential Program



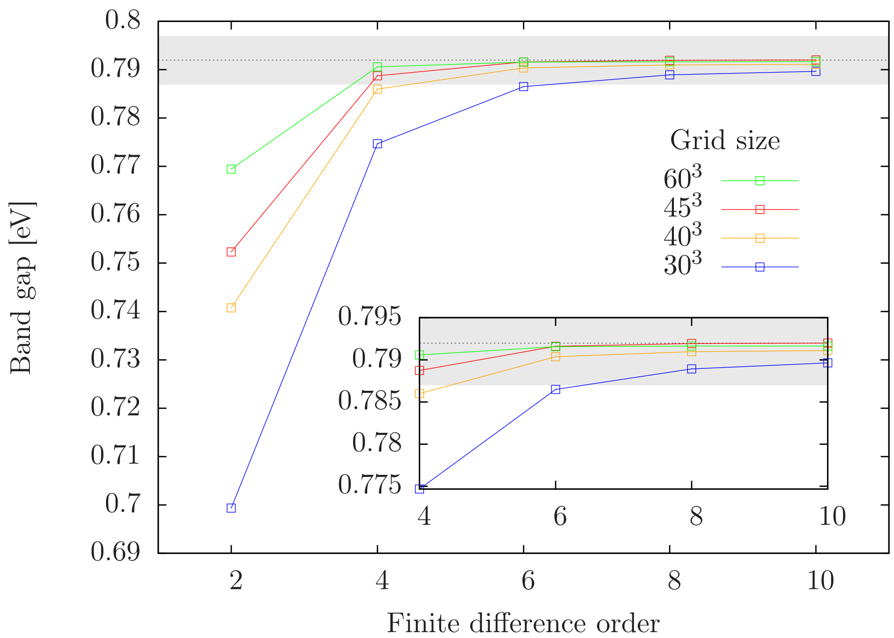
Real vs. reciprocal space convergence

Relation:

plane wave cut-off / real space grid points

$$E_{\mathrm{cut}} = \frac{1}{2}\left(\frac{N_d \pi}{a}\right)^2$$

$\Rightarrow$ common point of convergence



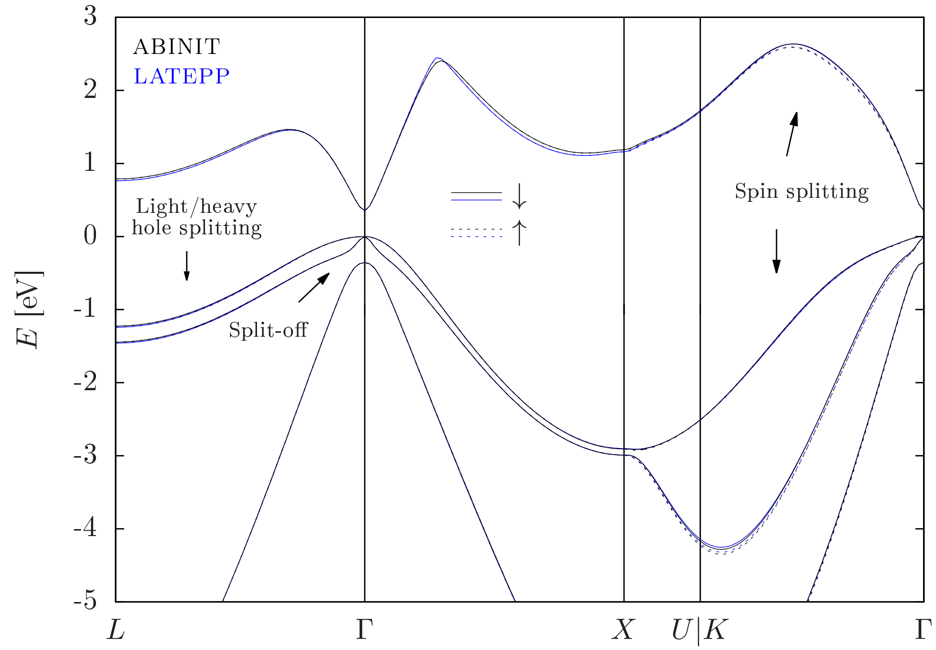
Finite difference order

Error ...

... reduces with increasing polynomial order

... decreases with increasing grid size

Large-Scale Atomic Effective Pseudopotential Program

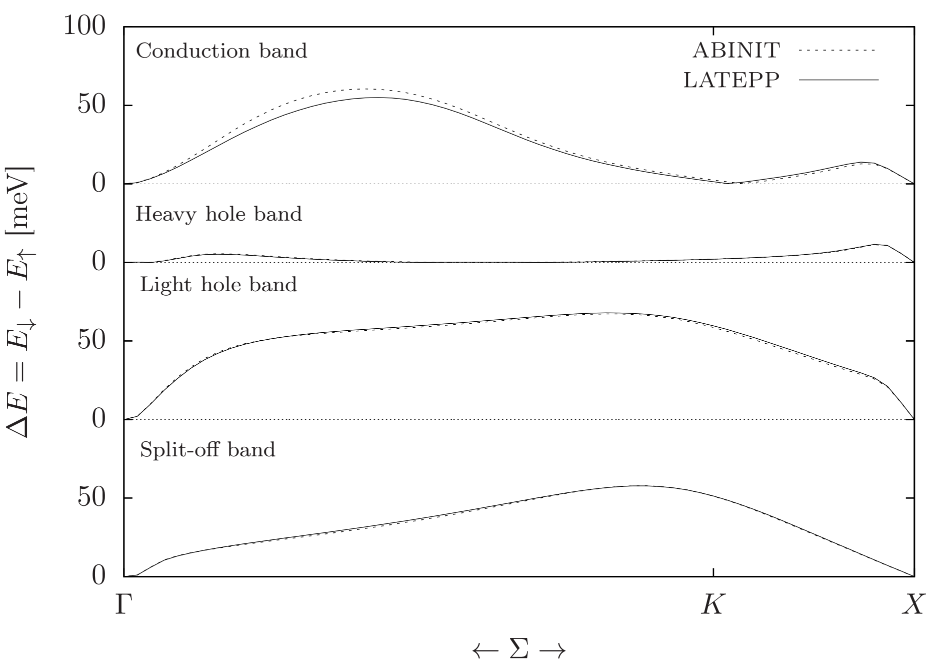


Spin-orbit treatment (in GaAs)

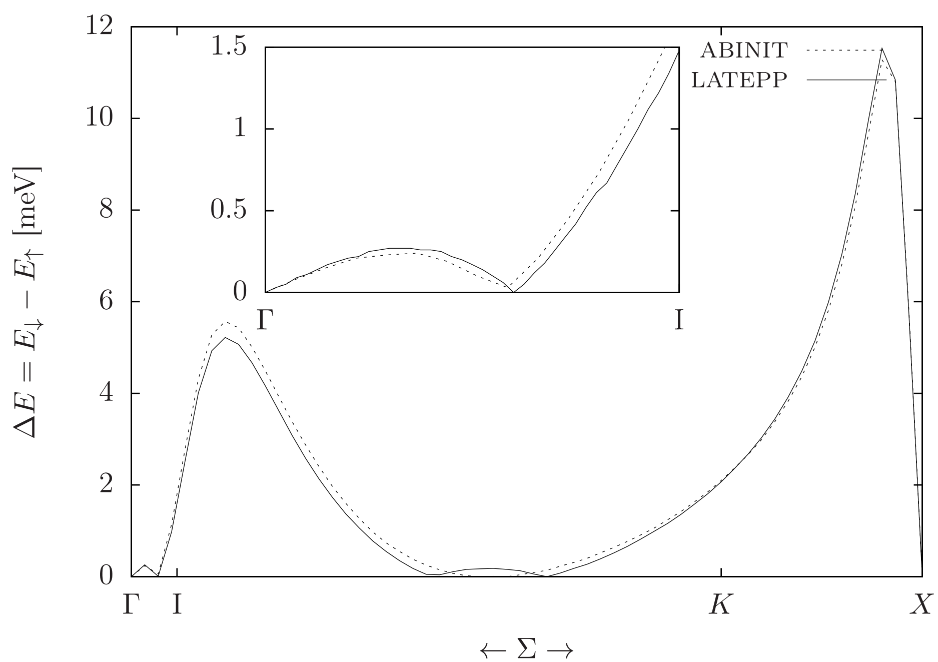
Efficiently evaluated in

- spin angular function basis
- Kleinman-Bylander formalism
- real space

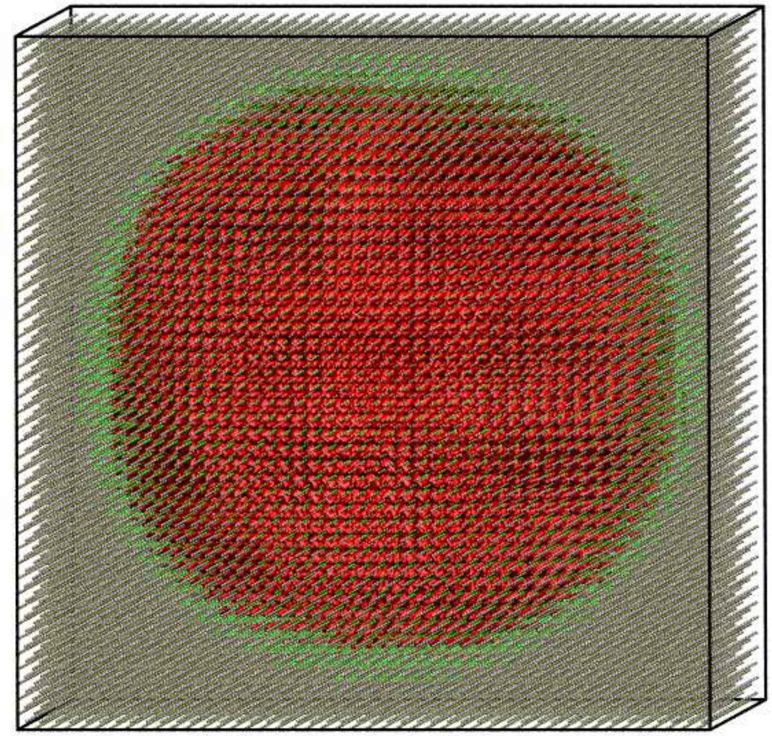
Perfect agreement to *ab initio* results



Heavy hole spin splitting



Summary — Conclusion — Outlook — Acknowledgements



LATEPP

Atomic
Effective
Pseudopotentials

+

Efficient iterative solver
Selected states of eigenspectrum

$\Rightarrow$ Large structures on *ab initio* level

Outlook

- *ab initio* quality wavefunctions
  $\rightarrow$ Configuration Interaction
- Beyond DFT: Correction of quasiparticle ...
  - effective masses: Nonlocal self-energy potential (empirical parametrization)
    $$H_{\mathrm{DFT}}\Psi_{n\boldsymbol{k}}(\boldsymbol{r}) + \int \Sigma(\boldsymbol{r},\boldsymbol{r}';E_{n\boldsymbol{k}})\Psi_{n\boldsymbol{k}}(\boldsymbol{r}')d\boldsymbol{r}' = E_{n\boldsymbol{k}}\Psi_{n\boldsymbol{k}}(\boldsymbol{r})$$
  - band gaps: Energy dependence of $\boldsymbol{\Sigma}$ — shifting eigenvalues by scissors operator
    $$E_{\mathrm{g}}^{\mathrm{LDA}} \longrightarrow E_{\mathrm{g}}^{\mathrm{LDA}} + \Delta$$

Acknowledgements

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Thank you for your attention!