



Parallel Empirical Pseudopotential Electronic Structure Calculations for Million Atom Systems

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We present a parallel implementation of the previously developed folded spectrum method for empirical pseudopotential electronic structure calculations. With the parallel implementation we can calculate a small number of electronic states for systems of up to one million atoms. A plane-wave basis is used to expand the wavefunctions in the same way as is commonly used in *ab initio* calculations, but the potential is a fixed external potential generated using atomistic empirical pseudopotentials. Two techniques allow the calculation to scale to million atom systems. First, the previously developed folded spectrum method allows us to calculate directly a few electronic states of interest around the gap. This makes the scaling of the calculation $O(N)$ for an N

the method has $O(N)$ scaling for a fixed number of states. Section II of this paper describes the computational aspects of the folded spectrum method in detail. This method has been applied to the study of free-standing [8–11] and embedded [17] quantum dots, as well as to

state $\{\psi\}$ by seeking the variational minimum of

$$F = \langle \psi | (\hat{H} - \epsilon_{\text{ref}})^2 | \psi \rangle. \quad (3)$$

Unlike the variational minimum of $\langle \psi | \hat{H} | \psi \rangle$, which yields the lowest energy state of \hat{H} , the minimum solution of F gives the band edge states if ϵ_{ref} is placed inside the band gap. Although simple algorithmically, the real challenge of the FSM is to develop a scheme which finds ψ efficiently from the minimization of F . This is not an easy task because changing H to F significantly increases the condition number of the linear operator (the matrix) [21]. We have used the conjugate gradient method to solve the variational minimum of F (this is described in detail in Ref. [19]). The Lanczos method is also appropriate for this type of problem, and a study of a variant of the method applied to the FSM is presented in [22].

To explain how we use the conjugate gradient method for this problem we must go into more detail of the plane-wave expansion for the wavefunctions. In a plane-wave representation the wavefunctions can be written as

$$\psi(\mathbf{r}) = \sum_{\mathbf{g}} a(\mathbf{g}) e^{i\mathbf{g}\cdot\mathbf{r}}. \quad (4)$$

The selection of the number of plane-waves is determined by a cutoff E_{cut} in the plane-wave kinetic energy $\frac{1}{2}|\mathbf{g}|^2$, where $\{\mathbf{g}\}$ are the reciprocal lattice vectors. The wavefunction ψ is stored in reciprocal space by its coefficients $a(\mathbf{g})$. It is transformed onto a real space grid $\psi(\mathbf{r})$ by applying a parallel FFT, which will be described in the next section.

Application of $(\hat{H} - \epsilon_{\text{ref}})^2$ to ψ is carried out [19] by twice applying $[-\frac{1}{2}\nabla^2 + V(\mathbf{r}) - \epsilon_{\text{ref}}]$ to ψ . The term $-\frac{1}{2}\nabla^2\psi$ is computed in reciprocal space, while $V(\mathbf{r})\psi(\mathbf{r})$ is obtained by using an FFT to transform $a(\mathbf{g})$ to real space, $\psi(\mathbf{r})$, then applying $V(\mathbf{r})$ to $\psi(\mathbf{r})$ and transforming the product back to \mathbf{g} space. The result can be cast in the same form as $\sum_{\mathbf{g}} c(\mathbf{g})e^{i\mathbf{g}\cdot\mathbf{r}}$ (with the same energy cutoff for $\{\mathbf{g}\}$). Then $[-\frac{1}{2}\nabla^2 + V(\mathbf{r}) - \epsilon_{\text{ref}}]$ is applied again to this function to get the final result F . Once F is obtained, we minimize it with respect to the variational wavefunction coefficients $a(\mathbf{g})$, using the preconditioned conjugate gradient method [23]. The conjugate gradient method is defined as a series (indexed by superscript $\{j\}$) of sequential line minimizations of the task function F . A line minimization implies adding a search wavefunction

The preconditioner $A(\mathbf{g})$ is a \mathbf{g} -space function

$$A(\mathbf{g}) = \frac{E_k^2}{\left(\frac{1}{2}g^2 + V_0 - \epsilon_{\text{ref}}\right)^2 + E^{2k}}$$

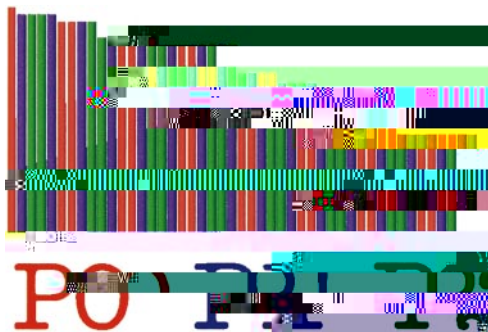


FIG. 2. Distribution of g vector columns to three processors. The g vector columns are produced by dividing the sphere of g vectors into z direction columns (see Fig. 3a). The columns are then ordered by length and assigned to the three processors as shown, with processor zero being assigned all the red columns, processor one the blue columns, and processor two the green columns.

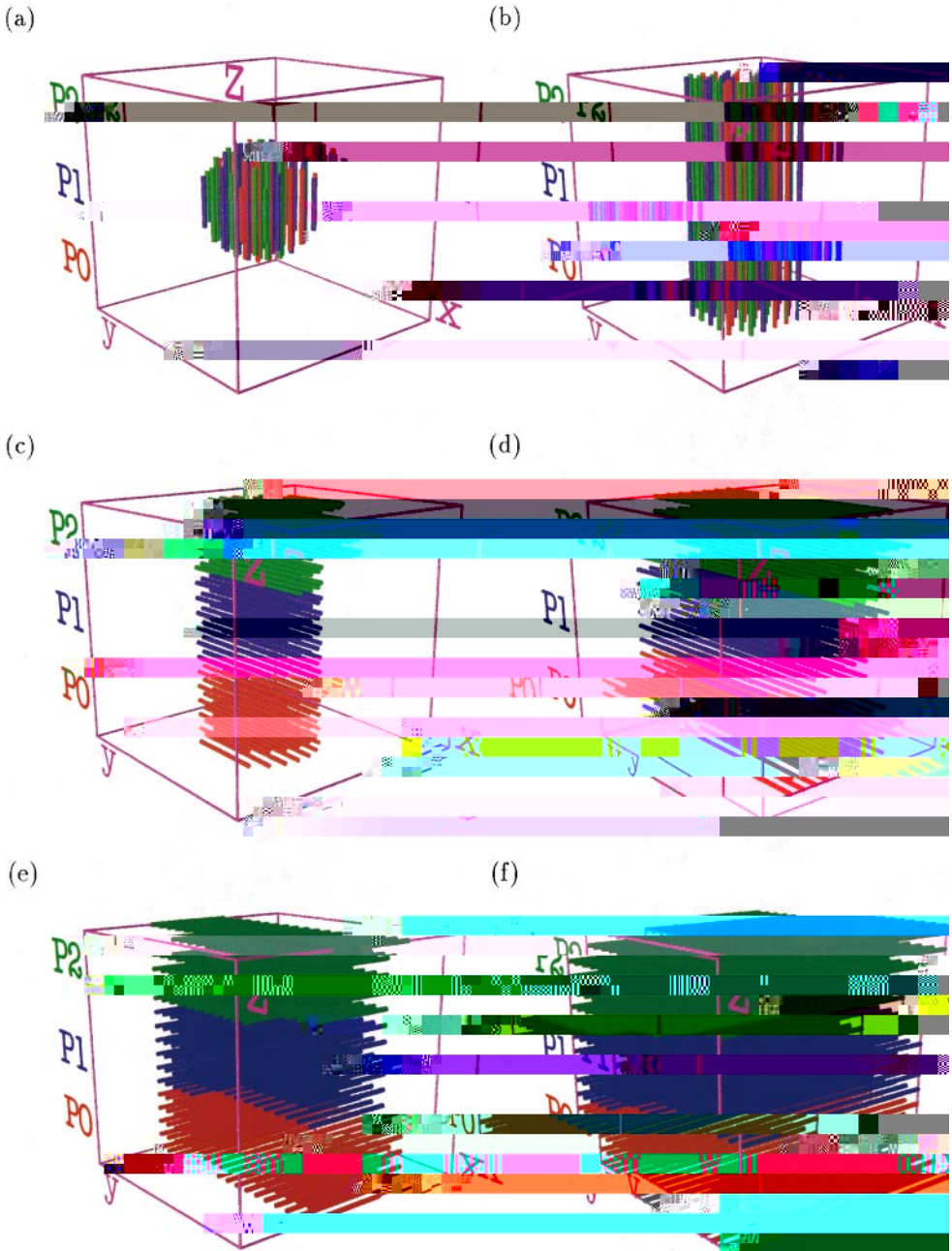


FIG. 3. Parallel three-dimensional FFT. This shows which processors deal with which part of the grid during the three dimensional FFT. The colors red, blue, and green correspond to the part of the grid that resides on processors zero, one, and two (for more details see text in Section III).

The size of the real space grid for the potential is typically taken to be twice the diameter of the g space sphere. A large saving in computations and communications can be made by not performing FFTs on the zero columns and not transforming the zero columns. At each step of the distributed 3d FFT, the data we are working on are expanding out to fill the full real space grid. For this reason it is inefficient to use FFT parallel libraries, which

cannot take advantage of this saving and often have very restricted data layouts such as

where $\{R_\alpha\}$ are the atomic positions of atom type α . The spherical atomic empirical pseudopotentials $v_\alpha(r)$ are obtained via a fit to the bulk band structure of the constituent materials [6]. The empirical pseudopotentials of Ref. [26] are used for InAs and GaAs. Using this method, the potential $V(\mathbf{r})$ of a million atom system can be readily constructed. The main task here is to calculate the wavefunctions $\psi_i(\mathbf{r})$ near the band edge of the energy spectrum (i.e., i near N_{occ}). A 5 Ryd cutoff energy is used for the plane-wave basis in Eq. (4).

To test the speed of the code for different numbers of processors and different system sizes we chose three InAs/GaAs quantum dot systems containing 8000, 97,336, and 1,000,000 atoms (see Fig. 4). The real space grid sizes for the potential for these systems are 128^3 ,

V. APPLICATION: ELECTRONIC STATES OF A PYRAMIDAL QUANTUM DOT

The ESCAN code is particularly suited to studying quantum dots, as it is reasonably straightforward to construct accurate empirical pseudopotentials for these systems, and the number of atoms, while being beyond the size possible with *ab initio* codes, is within

potential. As a result, the wavefunction $\psi_i(\mathbf{r})$ has spin-up and spin-down components. The relaxed InAs pyramid is placed inside a $28a \times 28a \times 30a$ GaAs matrix periodic supercell. The resulting system contains one quarter million atoms. We have calculated the four conduction band minimum (CBM) states and four valence band maximum (VBM) states. The real space grid size is $448 \times 448 \times 480$. On a 128-processor run, each processor holds 298 z -columns and holds on average 42,300 g coefficients for each wavefunction. The number of g coefficients varies from 42,284 to 42,310, which balances the memory usage on each processor. The whole calculation takes about 20 h on 128 processors of a Cray T3E computer.

The band gap increases from 0.41 eV for bulk InAs to 0.96 eV in the quantum dot. The charge density of the four CBM states and four VBM states are plotted in Fig. 5. They are all localized inside the quantum dot. A more detailed account of this work has been reported in [26].

VI. CONCLUSION

We have introduced an atomistic approach to calculating the electronic states of systems up to one million atoms. In this approach, the wavefunction is expanded using a plane-wave basis, as in conventional

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