

The use of bulk information to improve the scalability of parallel band gap computations for quantum dots

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Abstract. We consider the parallel computation of interior eigenstates of large Hermitian matrices arising from plane-wave discretization of effective single-particle Schrödinger equations.

We apply a folded-spectrum approach based on Preconditioned Conjugate Gradient (PCG) to compute only a small number of eigenstates close to the band gap whose location determines electronic and optical properties of the system.

We show how to improve the scalability of the eigensolver from observing that the quantum dot band states can be well approximated by states of bulk systems. We make use of these cheaply computable bulk eigenstates to improve the choice of the starting vector and the preconditioner for the eigensolver.

1 Introduction

The computation of electronic properties of large nano structures such as quantum dots is an important field of current research. Its significance is underlined by a number of activities such as the DOE-funded initiative "Predicting the Electronic Properties of 3D Million-Atom Semiconductor Nanostructure Architectures" that supports this current work.

We are interested in enhancing electronic structure calculations that are based on the solution of effective single-particle Schrödinger equations

$$H\Psi_i \equiv \left[-\frac{1}{2}\nabla^2 + V \right] \Psi_i = E_i\Psi_i. \quad (1)$$

In this equation, H denotes the approximate Hamiltonian. The atomic system is described by the potential V which we assume as externally given empirical pseudopotential [2]. If the system has an electron with energy E_i , then the wave function $\Psi_i(r)$ describes the spatial probability distribution for the electron.

The physical interest often lies in only a small number of eigenstates of (1) which are relevant for determining certain optical and electronic properties. A common task is to compute the states at the top of the valence and at the bottom of the conduction band to find the band gap.

In a discrete finite plane-wave basis, equation (1) directly translates into a Hermitian eigenvalue problem and we use the same notation H for the matrix in question. H is only implicitly available through matrix vector products and via the Fast Fourier Transform (FFT). This together with the system size requires the use of efficient iterative eigenvalue methods.

Our parallel Energy SCAN (ESCAN) method [2] uses a folded spectrum approach [6] to find interior eigenstates. Based on physical knowledge about the system, a reference energy E_r is chosen and then the smallest eigenvalues of the system $(H - E_r)^2$ are computed via Preconditioned Conjugate Gradient(PCG) Rayleigh-Quotient minimization, see the references in [4] and [5]. However, as described in Section 2, there can be convergence problems for large systems with strongly clustered, nearly degenerate eigenstates. The squaring of the matrix also contributes to the stronger clustering of the eigenvalues.

2 PCG for quantum dot valence band computations

The smallest eigenvalue λ of the Hermitian matrix $A \equiv (H - E_r)^2$ (the one that corresponds to the eigenvalue of H closest to the reference point E_r) minimizes the Rayleigh Quotient, that is

$$\lambda = \arg \min_{x \neq 0} \rho(x) \equiv \frac{x^* Ax}{x^* x}. \quad (2)$$

From a current iterate x_k and a descent direction p_k , the method finds the angle

$$\theta_k = \arg \min \rho(x_{k+1}) \equiv \rho(x_k \cos \theta_k + p_k \sin \theta_k). \quad (3)$$

The descent direction is given by $p_k = -\nabla \rho(x_k) + \beta_k p_{k-1}$, where the gradient $\nabla \rho(x) = 2(Ax - \rho(x)x)/(x^* x)$ is just a scalar multiple of the residual and β_k is a scalar for whose choice various strategies exist, see [4]. A preconditioner P can be used to influence the choice of the descent direction [3] via

$$p_k = -P \nabla \rho(x_k) + \beta_k p_{k-1}. \quad (4)$$

As shown later, the preconditioner in the current version of ESCAN [2] can lead to very slow convergence in valence band computations of quantum dots. In the following, we discuss possibilities to improve this.

3 How to make use of bulk information

The properties of ideal bulk systems such as crystals are well understood by material scientists. Each so-called Bloch eigenstate of the bulk Hamiltonian is of the form $\Psi_{nk}(r) = u_{nk}(r) * \exp(-ik * r)$, where $u_{nk}(r)$ is periodic [1].

In contrast, quantum dots represent more complicated physical objects where bulk materials constitute the interior and vacuum the exterior. However, the key observation on which we base our approach is that for large enough systems, the converged quantum dot states around the band gap have a small angle to the subspace defined by the corresponding bulk system states [7].

3.1 An example

As an example, we consider a bulk system and a quantum dot consisting of Cadmium (Cd) and Selenium (Se) material. The characteristics of both systems are compared in Table 1.

| | bulk system | quantum dot |
|-------------------------|-------------|----------------|
| number of atoms | 1 Cd, 1 Se | 784 Cd, 739 Se |
| dimension(H) plane wave | 2048 | 1.45E+5 |

Table 1. Comparison between bulk and quantum dot.

3.2 Selection of an improved initial vector

While the Rayleigh-Ritz procedure is too expensive, we can use the bulk subspace to find a good initial vector for the PCG iteration. Experimentally, we found that the corresponding bulk wave function at the gamma point (the center of the first Brioullin zone [1]). constitutes an excellent starting vector for the PCG iteration.

We use the gamma point bulk state Ψ_{n0} and limit its support to the interior of the quantum dot, that is we cut off the initial vector in the part that corresponds to vacuum. From physics, it is known that the true solution we are looking for typically is confined to the interior of the quantum dot, see the picture on the left of Figure 1.

3.3 The previously used preconditioner

The preconditioner that was previously used in ESCAN is the diagonal

$$P = D \equiv (I + (-\frac{1}{2}\nabla^2 + V_{avg} - E_{ref})/E_k)^2$$

where $-\frac{1}{2}\nabla^2$ is the Laplacian (diagonal in the Fourier space), E_{ref} is the shift used in the folded spectrum, V_{avg} is the average potential and E_k is the average kinetic energy of a given initial approximation of a wave function ψ_{init} .

3.4 An improved preconditioner based on bulk band computation

We define H_{BB} to be the Hamiltonian stemming from the discretization of the Schrödinger's equation for a bulk system. Denote its eigenfunctions by $\Psi_{nk}(r)$ with corresponding energies E_{nk} .

We decompose the residual R into its S_{BB} and S_{BB}^\perp components, i.e. $QQ^T R$ and $R - QQ^T R$, and precondition the S_{BB} component with H_{BB}^{-1} and the rest with the diagonal preconditioner D^{-1} , i.e.

$$PR \equiv QH_{BB}^{-1}Q^T R + D^{-1}(R - QQ^T R). \quad (5)$$

3.5 Comparison of results

Figure 1 shows the improvements over the old preconditioner with random initial vector. From top to bottom, see the curves for the old preconditioner with random initial vector, old preconditioner with improved initial vector, and new preconditioner with improved initial vector.

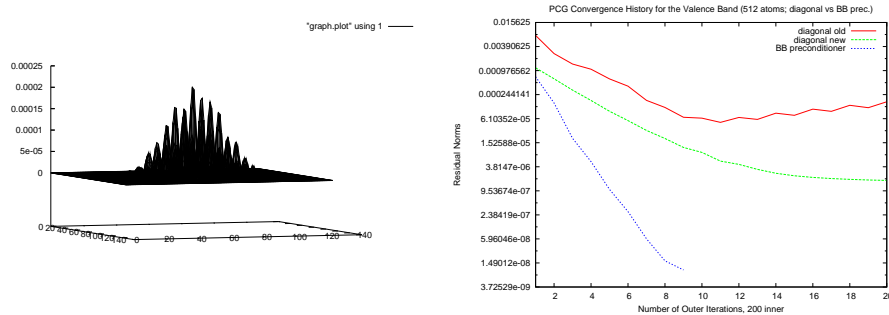


Fig. 1. *Left hand side:* cross section of the charge density for the state at the top of the valence band. The wave-function is clearly confined to the interior of the quantum dot. *Right hand side:* Convergence for computation of the corresponding state. The new initial vector improves the convergence of the old diagonal preconditioner. The new preconditioner together with the new initial vector further improves the convergence.

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