Graphics Processing Unit Acceleration of the Random Phase Approximation in the Projector Augmented Wave Method

Jun Yan\textsuperscript{1,*}, Lin Li\textsuperscript{1}, Christopher O’Grady\textsuperscript{1}

\textit{SUNCAT Center for Interface Science and Catalysis, SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, CA 94025, USA}

Abstract

The Random Phase Approximation (RPA) for correlation energy in the grid-based projector augmented wave (GPW) code is accelerated by porting to the Graphics Processing Unit (GPU) architecture. The acceleration is achieved by grouping independent vectors/matrices and transforming the implementation from being memory bound to being computation/latency bound. With this approach, both the CPU and GPU implementations have been enhanced. We tested the GPU implementation on a few representative systems: molecules (O\textsubscript{2}), bulk solids (Li\textsubscript{2}O and MoO\textsubscript{3}) and molecules adsorbed on metal surfaces (N\textsubscript{2}/Ru(0001) and CO/Ni(111)). Improvements from 10× to 40× have been achieved (8-GPUs versus 8-CPUs). A realistic RPA calculation for CO/Ni(111) surface can be finished in 5.5 hours using 8 GPUs. It is thus promising to employ non-self-consistent RPA for routine surface chemistry simulations.

Keywords: Graphics Processing Unit, Adiabatic connection fluctuation-dissipation theory, Random Phase Approximation

1. Introduction

Density functional theory (DFT) has became one of the standard tools for predicting the structural, energetic, mechanical, dielectric and magnetic properties of condensed matter. In the field of computational catalytic design, thousands of DFT calculations have been reported and tabulated for different adsorption species on a variety of metal surfaces with different facets \[1\]. The general trend of the variations in catalytic activity from one catalyst to another is reported to be well captured by DFT calculations \[2\]. However, DFT does not yet achieve the chemical accuracy (1 kcal/mol or 0.0434 eV) needed to predict

\*Corresponding author

Email address: junyan@stanford.edu (Jun Yan)
the energetics of many catalysts. This level of accuracy is important because the reactivity depends exponentially on adsorption and reaction energies. Furthermore, for systems with strong correlation and localization, it is unclear if DFT can qualitatively predict the correct catalytic trends.

Random Phase Approximation (RPA) has emerged as a promising approach to improve the precision of total energy predictions in computational chemistry and materials science \[3, 4, 5\]. Instead of using the local or semi-local exchange-correlation functionals of standard DFT calculations, RPA accounts for dynamic electronic screening and is fully non-local. It has been shown to systematically improve lattice constants \[6\], atomization and cohesive energies \[7\], adsorption sites and energies \[3, 4, 10\], reaction barriers \[3\], and structural phase transitions \[11\] for a wide range of systems that have ionic, covalent and/or van der Waals interactions \[12, 13, 14, 15\]. Similar to many other beyond-DFT calculations, the RPA method is exceptionally computational demanding since it involves summations over large basis sets and hundreds or thousands of unoccupied orbitals, both of which are truncated above a certain energy.

The most demanding part of an RPA calculation is to compute the non-interacting response function. There are a few methods proposed in the literature to speed up the calculation of the response function. These methods focus on reducing or eliminating the number of unoccupied orbitals, and have been applied to compute the self-energies in GW calculations \[16, 17\]. However, GW self-energies are known not to depend strongly on the approximations made to the imaginary part of the response function. It is unknown how such approximations would affect RPA calculations. A more rigorous algorithm using many-body perturbation theory can completely eliminate the unoccupied orbitals \[18, 19, 20\], however the algorithm requires solving a complex eigenvalue equation (either by diagonalizing or using a Lanczos scheme), and the number of basis functions used cannot be reduced. Recently, Bruneval proposed a range-separated approach: preserving the RPA long-range non-local correlation (which is the essence of the RPA method and is fast to converge in reciprocal space) and using a local functional to replace the short range correlation \[21\]. Such an approach significantly speeds up the convergence with respect to the number of states and has been applied to a silicon vacancy in a 216 atom supercell.

Graphics processing units (GPUs) are becoming attractive platforms for high performance scientific computing. A GPU contains hundreds of cores, with low price to performance ratio and a relatively low energy consumption per core compared to traditional CPUs. They are most advantageous for parallelizable algorithms requiring a large number of numerical operations per memory fetch (“numerically-intensive”). An increasing number of scientific applications have been ported to GPUs \[22\]. In particular, for electronic structure calculations, GPUs have been utilized for many quantum chemistry and DFT codes such as GPAW \[23\], VASP \[24, 25\], QUANTUM ESPRESSO \[26\], TERACHEM \[27\], BIGDFT \[28\], PETOT \[29\] and OCTOPUS \[30\]. The speed up of these DFT-based electronic structure codes is generally less than 15× due to the complexity and communication bottlenecks in algorithms such as FFT, subspace diagonaliza-
tion, minimization and orbital orthogonalization. These problems, however, do not apply to beyond DFT methods based on the linear density response function such as the RPA correlation energy. The evaluation of the linear density response matrix $\chi^0_{GG'}(q, \omega)$ is in general numerically-intensive. The single particle transitions that are required to calculate the response matrix are completely independent of each other. This allows for straightforward MPI parallelization, with no MPI communication required during the calculation. It is thus natural to port the RPA method to GPUs.

This paper describes the GPU porting of the RPA method (specifically, the evaluation of the linear density response function) as well as the performance on a few representative molecules, bulk solids and surfaces. The rest of the paper is organized as follows. In Section 2 the theory and algorithm of RPA is briefly reviewed, focusing on the CPU implementation. Section 3 presents a simpler “direct” GPU porting, which is achieved with minimal changes to the code structure, followed by a “multi-u” technique that enhances both the GPU and CPU implementations. The performance of the GPU implementation for a few representative systems is discussed in Section 4 and finally conclusions are given in Section 5.

2. The RPA Method and the CPU Implementation

The RPA scheme for obtaining the total energy consists of two parts: exact exchange energy and correlation energy using RPA. Both can be derived from the adiabatic connection fluctuation-dissipation theory (ACFDT)\cite{31}. Here we only briefly review the portion of RPA theory that is relevant to the GPU porting of the code, and focus instead on the actual implementation. The theory on the exact exchange energy and its porting to the GPU platform can be found in Ref. \cite{32}.

2.1. Correlation energy using random phase approximation

According to ACFDT, the RPA correlation energy $E_{rpa}^c$ can be formulated as

$$E_{rpa}^c = \int_0^\infty \frac{d\omega}{2\pi} Tr\{ln[1 - v\chi^0(i\omega)] + v\chi^0(i\omega)\},$$

(1)

where $v$ is the Coulomb interaction kernel and $\chi^0$ is the non-interacting response function. $\chi^0$ is a fundamental quantity in many beyond-DFT methods and is described in the following section.

In the GPAW \cite{33, 34}, RPA implementation \cite{35}, $v$ and $\chi^0$ are represented using a plane wave basis set and become $v_G(q)$ and $\chi^0_{GG'}(q, i\omega)$, respectively. $q$ is a wave vector within the Brillouin zone (BZ) and $G$ is a reciprocal space lattice vector, the size of which is defined by a cutoff energy $E_{cut}$. The RPA correlation energy under the plane wave representation becomes

$$E_{rpa}^c = \int_0^\infty \frac{d\omega}{2\pi} \int_{BZ} dq Tr\{ln[1 - v_G(q)\chi^0_{GG'}(q, i\omega)] + v_G(q)\chi^0_{GG'}(q, i\omega)\}$$

(2)
The frequency integration over $\omega$ is carried out using 16 Gauss-Legendre points following the procedure from Ref. [13]. The integration over the BZ is discretized using Monkhorst-pack $k$-points. By exploiting the $q$-mesh symmetry, the integration is reduced to a summation over the irreducible BZ (IBZ) : $\int_{BZ} \rightarrow \sum_{IBZ} w_q$ where $w_q$ is the weight for a specific $q$ vector.

2.2. Density response function in the projector-augmented wave method

The non-interacting response function is evaluated in a plane wave basis using the “sum over states” approach written as

$$\chi^0_{G\bar{G}'}(q, i\omega) = \frac{2}{\Omega} \sum_{k} \sum_{n,n'} \int \frac{f_{nk} - f_{n'k+q}}{i\omega + \epsilon_{nk} - \epsilon_{n'k+q}} n_{nk,n'+k+q}(G)n_{nk,n'+k+q}'(G')$$

(3)

where

$$n_{nk,n'+k+q}(G) \equiv \langle \psi_{nk}| e^{-(q+G) \cdot r} | \psi_{n'k+q} \rangle$$

(4)

is the charge density matrix and $\Omega$ is the volume of the unit cell. The occupation $f_{nk}$, Kohn-Sham (KS) eigenvalue $\epsilon_{nk}$ and eigenstate $\psi_{nk}$ for band $n$ at wave vector $k$ are extracted from a DFT calculation. The spin index $\sigma$ is implicitly contained in the above formula.

In the PAW formalism [30], a true all-electron KS wave function $\psi_{nk}$ is obtained by a linear transformation $T$ from a smooth pseudo-wave function $\tilde{\psi}_{nk}$ via $\psi_{nk} = T \tilde{\psi}_{nk}$. The transformation operator is chosen in a manner such that the all electron wave function $\psi_{nk}$ is the sum of the pseudo one $\tilde{\psi}_{nk}$ plus a contribution centered around each atom written as

$$\psi_{nk}(r) = \tilde{\psi}_{nk}(r) + \sum_{a,i} \langle \tilde{p}^a_i \psi_{nk} | \phi^a_i(r - R_a) - \tilde{\phi}^a_i(r - R_a) \rangle$$

(5)

The pseudo-wave function $\tilde{\psi}_{nk}$ matches the all-electron one $\psi_{nk}$ outside the augmentation spheres centered on each atom $a$ at position $R_a$. Their differences inside the augmentation spheres are expanded on atom-centered all-electron partial waves $\phi^a_i$ and the smooth counterparts $\tilde{\phi}^a_i$. The expansion coefficient is given by $\langle \tilde{p}^a_i | \psi_{nk} \rangle$, where $\tilde{p}^a_i$ is a dual basis to the pseudo-partial wave and is called a projector function. In the practical GPAW implementation, the pseudo-wave function can be discretized using plane waves, three-dimensional uniform real space grids, or a localized atomic orbital basis. The atomic centered quantities such as $\tilde{p}^a_i$, $\phi^a_i$ and $\tilde{\phi}^a_i$ are expanded using a one dimensional logarithmic grid, which becomes denser closer to the atomic cores.

Substituting Eq. (5) into Eq. (4), the charge density matrix becomes

$$n_{nk,n'+k+q}(G) = \tilde{n}_{nk,n'+k+q}(G) + \sum_{a,i,j} \langle \tilde{\psi}_{nk} | \tilde{p}^a_i | \psi_{n'k+q} \rangle Q_{ij}^a (q + G)$$

(6)

with definitions

$$\tilde{n}_{nk,n'+k+q}(G) \equiv \langle \tilde{\psi}_{nk}| e^{-(q+G) \cdot r} | \tilde{\psi}_{n'k+q} \rangle$$

(7)
\[ Q_{ij}^\alpha(q + G) = \langle \phi_i^\alpha | e^{-i(q+G) \cdot \mathbf{r}} | \phi_j^\alpha \rangle - \langle \tilde{\phi}_i^\alpha | e^{-i(q+G) \cdot \mathbf{r}} | \tilde{\phi}_j^\alpha \rangle. \]  

In the above equations, \( \mathbf{k} \) and \( \mathbf{k} + q \) are wave vectors within the BZ; however in a general DFT calculation the eigenvalues and eigenstates are computed only for \( k \)-points in the IBZ. As a result, a mapping of the \( k \)-point indices from BZ to IBZ is required:

\[ \mathbf{k} = T_1 \mathbf{k}_{1IBZ}, \mathbf{k} + q = T_2 \mathbf{k}_{2IBZ}, \]

where \( T_1 \) and \( T_2 \) are the transformation operators for \( \mathbf{k} \) and \( \mathbf{k} + q \), respectively. Correspondingly, a transformation of the KS eigenvectors from IBZ to BZ using crystal symmetry operations is needed and will be described in the following.

For the RPA implementation, the pseudo-wave function is expanded using a plane wave basis. This choice is motivated by the fact that a typical RPA correlation energy calculation requires hundreds to thousands of eigenstates, which can be efficiently computed by direct diagonalization of the KS Hamiltonian in a plane wave basis using a previously converged self-consistent DFT calculation. Given plane wave coefficients \( C_{nk1}^{IBZ}(Q) \) and \( C_{nk2}^{IBZ}(Q') \), where \( Q \) and \( Q' \) are reciprocal lattice vectors, the pseudo-wave function in the IBZ is obtained through

\[ \tilde{\psi}_{nk}^{IBZ}(\mathbf{r}) = \sum_Q C_{nk1}^{IBZ}(Q) e^{i(k_{1IBZ} + Q) \cdot \mathbf{r}}, \tilde{\psi}_{n'k}^{IBZ}(\mathbf{r}) = \sum_{Q'} C_{n'k2}^{IBZ}(Q') e^{i(k_{2IBZ} + Q') \cdot \mathbf{r}}, \]

which can be performed efficiently using a Fast Fourier Transform (FFT). The transformation of the pseudo-wave function from IBZ to BZ is

\[ \tilde{\psi}_{nk}(\mathbf{r}) = \tilde{\psi}_{nk}^{IBZ}(T_1^{-1} \mathbf{r}), \tilde{\psi}_{n'k+q}(\mathbf{r}) = \tilde{\psi}_{n'k}^{IBZ}(T_2^{-1} \mathbf{r}). \]

Both crystal symmetries and time-reversal symmetry are taken into account in the above transformation.

After obtaining the \( \tilde{\psi}_{nk}(\mathbf{r}) \) and \( \tilde{\psi}_{n'k+q}(\mathbf{r}) \) on the uniform 3D grid, the pseudo-density matrix \( \tilde{n}_{nk,n'k+q}(\mathbf{G}) \) is obtained using an FFT \( \mathcal{F} \) through

\[ \tilde{n}_{nk,n'k+q}(\mathbf{G}) = \mathcal{F}[\tilde{\psi}_{nk}^* (\mathbf{r}) \tilde{\psi}_{n'k+q}^* (\mathbf{r}) e^{-i \mathbf{q} \cdot \mathbf{r}}]. \]

Compared to directly integrating the wave functions for each \( \mathbf{G} \) on the 3D grid, the use of an FFT achieves a speed up \( > 100 \times \).

For the augmentation part in Eq. 4, the \( \langle \tilde{\psi}^p_{nk} | \tilde{\psi}_{nk}^{IBZ} \rangle \) and \( \langle \tilde{\psi}^p_{nk} | \tilde{\psi}_{n'k}^{IBZ} \rangle \) are calculated and saved during DFT calculations for \( k \)-points that are inside the IBZ. For \( k \)-points outside the IBZ, symmetry operations are applied to the projector function \( \tilde{\psi}^p_{nk} \) and \( \tilde{\psi}^p_{n'k} \) to obtain \( \langle \tilde{\psi}^p_{nk} | \tilde{\psi}_{nk} \rangle \) and \( \langle \tilde{\psi}^p_{nk} | \tilde{\psi}_{n'k+q} \rangle \), respectively. The \( Q_{ij}^a(q + G) \) is calculated on a 1D logarithmic grid by expanding the \( e^{-i(q+G) \cdot \mathbf{r}} \) using a spherical harmonic basis. Since it requires only a single shot calculation, it is performed at the initialization step of an RPA calculation on the CPU and then copied to the GPU.

For the case \( q = 0 \) and \( G = 0 \), the charge density matrix in Eq. 4 becomes \( \langle \tilde{\psi}^n_{nk} | \tilde{\psi}_{nk} \rangle = \delta_{nn'} \); on the other hand, the coulomb kernel \( \nu_G(q) = 4\pi / |q + G|^2 \) becomes divergent. To cure the divergence of the coulomb kernel, a perturbative
approach is used by taking the $q \rightarrow 0$ limit and the charge density matrix is calculated using

$$n_{q \rightarrow 0}(G = 0) = \langle \psi_{nk} | e^{-iQ \cdot \mathbf{r}} | \psi_{n'k+q} \rangle_{q \rightarrow 0}$$

(13)

$$= -iQ \cdot \langle \psi_{nk} | \nabla | \psi_{n'k} \rangle \overline{\epsilon_{n'k} - \epsilon_{nk}}$$

(14)

In the above derivation, $\psi_{n'k+q}$ at $q \rightarrow 0$ is expanded using $k \cdot p$ second order perturbation theory [35]. In the PAW method, the matrix element $\langle \psi_{nk} | \nabla | \psi_{n'k} \rangle$ is given by

$$\langle \psi_{nk} | \nabla | \psi_{n'k} \rangle = \langle \tilde{\psi}_{nk} | \nabla | \tilde{\psi}_{n'k} \rangle + \sum_{a,ij} \langle \tilde{\psi}_{nk} | \tilde{p}_a^j | \tilde{\psi}_{n'k} \rangle Q^a_{ij}(G = 0)_{q \rightarrow 0}$$

(15)

with

$$Q^a_{ij}(G = 0)_{q \rightarrow 0} = \langle \tilde{\phi}_i^a | \nabla | \tilde{\phi}_j^a \rangle - \langle \tilde{\phi}_j^a | \nabla | \tilde{\phi}_i^a \rangle$$

(16)

The pseudo-wave function part $\langle \tilde{\psi}_{nk} | \nabla | \tilde{\psi}_{n'k} \rangle$ is calculated using a finite difference approximation for the nabla operator, taking into account 6 neighboring points (in total 13 grid points) in each direction. The $Q^a_{ij}(G = 0)_{q \rightarrow 0}$ is calculated by expanding the partial waves ($\tilde{\phi}_i^a$, $\tilde{\phi}_j^a$) on real spherical harmonics and applying the nabla operator on the radial and angular part of the expansion coefficients. For a detailed derivation of the $Q^a_{ij}(G = 0)_{q \rightarrow 0}$ limit on a 1D logarithmic grid, refer to Ref. [35].

2.3. The CPU implementation flow

The response function $\chi^0_{GG'}(q, i\omega)$ in Eq. (3) is implemented as a double precision matrix of size $(nG)^2 \times nq \times n\omega$, where $nG, nq, n\omega$ corresponds to the number of plane waves, q-points and frequency points, respectively. The sizes of $nG$ and $nq$ are system dependent, and are typically a few hundreds to thousands, and a few tens to hundreds, respectively. $n\omega$ corresponds to the 16 Gauss-Legendre points. Considering that a typical memory of 2-3 Gigabytes is available per core, we choose to loop over q-points and store the matrix $\chi^0_{GG'}(i\omega)$ in memory during computations.

In order to obtain $\chi^0_{GG'}(i\omega)$ at a given $q$, we need to compute the charge density matrix $n_{nk,n'k+q}(G)$ in Eq. (4). The number of bands (index $n$ and $n'$) and number of k-points (index $k$) are generally too large, so the entire density matrix can not reside in memory for RPA calculations. As a result, the computation of $\chi^0_{GG'}(i\omega)$ is achieved by looping and summing over $n$, $n'$, $k$ and $s$ (spin, implicitly included) indices, and calculating the charge density matrix, which is in fact a vector $n(G)$ of length $nG$, within each loop according to

$$\chi^0_{GG'}(i\omega) = \sum_{k,n,n'} A(i\omega)n(G)n^*(G'),$$

(17)

where $A(i\omega)$ is a vector defined as (given $n$, $n'$, $k$ and $q$)

$$A(i\omega) = \frac{2}{\Omega} \times \frac{f_{nk} - f_{n'k+q}}{i\omega + \epsilon_{nk} - \epsilon_{n'k+q}}$$

(18)
Table 1: RPA algorithm

1) Initialization, including MPI initialization and distribution;
2) Read plane wave coefficients $C_{nk^{IBZ}}(Q)$ and $C_{n'k'^{IBZ}}(Q')$, where $k^{IBZ}_1$ and $k^{IBZ}_2$ are the corresponding IBZ $k$-points for $k$ and $k + q$, respectively, following Eq. (9);
3) Calculate (using FFT) the pseudo-wave function $\tilde{\psi}_{nk^{IBZ}_1}(r)$ and $\tilde{\psi}_{n'k'^{IBZ}_2}(r)$ from the coefficients $C_{nk^{IBZ}_1}(Q)$ and $C_{n'k'^{IBZ}_2}(Q')$, respectively, following Eq. (10);
4) Transform the pseudo-wave function from IBZ to BZ and obtain $\tilde{\psi}_{nk}(r)$ and $\tilde{\psi}_{n'k'+q}(r)$ according to Eq. (11);
5) Calculate pseudo density matrix using FFT according to Eq. (12);
6) Map the FFT result on the FFT grid to a reduced grid $G$, the size of which is defined by the cutoff energy of the response function, to get $\tilde{n}(G)$;
7) Read $(\tilde{p}^a_i | \tilde{\psi}_{nk^{IBZ}_1})$ and $(\tilde{p}^a_i | \tilde{\psi}_{n'k'^{IBZ}_2})$, transform them from IBZ to BZ to obtain $P(a, i) \equiv (\tilde{p}^a_i | \tilde{\psi}_{nk})$ and $P(a, j) \equiv (\tilde{p}^a_i | \tilde{\psi}_{n'k'+q})$, respectively;
8) Perform $P(a, p) \equiv P^*(a, i) \otimes P(a, j)$, where $p \equiv \{ij\}$ is a combined index of $ij$;
9) Perform $\tilde{n}(G) + = \sum_{ap} P(a, p)Q(a, p, G)$. Steps 8) and 9) follow Eq. (13) - (14); otherwise skip this step;
10) If $q = 0$, calculate and replace $n(G = 0)$ using Eq. (13) - (14); otherwise compute the contribution to $E_{cRPA}$ at the particular $q$ according to Eq. (2); Gaussian;
11) Compute the contribution to $E_{cRPA}$ at the particular $q$ according to Eq. (2); Gaussian;
12) Steps 2 - 11 are looped over $n, n', k$ and $s$ (spin, not explicitly written) indices until the calculation of $\chi^0$ at a particular $q$ is finished;
13) Steps 12 - 13 are looped over $q$ until $E_{cRPA}$ is finished.

Table 1 shows the algorithm used to compute an RPA correlation energy. In order to make the size of the matrix clearer, we change the notation for the matrices in Table 1 so that the values inside the parentheses correspond to the size of the matrix. For instance, $C_{nk^{IBZ}_1}(Q)$ is a vector of length $NQ$. It is, however, slightly different for PAW related functions because the number of projector functions $P^a_i$ is different for each atom. For example, $P(a, i)$ represents a list of atoms of length $Na$ and for each atom, a vector of size $Ni$; while $Q(a, p, G)$ represents a list of atoms of length $Na$ and for each atom, a matrix of size $Np \times NG$.

Steps 2 - 11 take more than 99.9% of the total computing time and we focus on this part. Step 2 performance is determined by the I/O speed of reading orbitals from a previously saved DFT calculation. Step 3 is separated into two parts: first mapping the coefficient to the FFT grid and then performing the FFT. The transformation in step 4 corresponds to a mapping from one $r$ grid to another: $r' = T^{-1}r$. Step 5 contains two parts: evaluating...
\[ \tilde{\psi}_{n,k}^*(\mathbf{r})\tilde{\psi}_{n',k+\mathbf{q}}(\mathbf{r})e^{-i\mathbf{q} \cdot \mathbf{r}} \] on the 3D uniform real space grid and performing a 3D FFT. Step 6 maps the result on the 3D FFT grid to a reduced \( \mathbf{G} \) 1D grid. Steps 7 - 9 calculate the PAW corrections to the response function, with steps 8 and 9 corresponding to an outer and inner product of two functions for each atom, respectively, and finally a sum over atoms. Step 10 calculates the optical limit correction to the response function at \( \mathbf{G} = 0 \). Step 11 performs an outer product of two vectors. The BLAS library is exploited in step 8 (GEMM), step 9 (GEMV), and step 11 (ZHER), while the FFTW library is used for FFTs in steps 3 and 5.

3. Porting the RPA code to GPUs

Here we present two separate steps in the GPU porting process. A “direct” approach which makes minimal changes to the code, and a “multi-\( n \)” approach that uses higher performance computation/latency bound algorithms instead of memory bound algorithms, but requires more changes to the code structure.

3.1. Direct GPU porting

Direct GPU porting includes: replacing all the MKL BLAS function calls with CUBLAS and FFTW (version 3) with CUFFT (CUDA 5.0), as well as implementing a few cuda kernels such as wave function transformation, index mapping and and PAW projection evaluation. Double and double complex precision is used for both the CPU and GPU code. For each step, the GPU code maintains the same data structure and flow as the CPU code, except for step 10. The optical limit calculation for step 10 is performed using a finite difference method (the so called “stencil” method) for the derivative operator in Eq. (15). Instead of writing a cumbersome stencil kernel using CUDA, we reformulated the problem using an FFT. Given that the pseudo-wave function can be expanded in plane waves according to Eq. (10), one can write

\[ \nabla \tilde{\psi}_{n'k}(\mathbf{r}) = \nabla \left[ \sum_{\mathbf{Q}} C_{n'k}(\mathbf{Q}) e^{i(\mathbf{k}+\mathbf{Q}) \cdot \mathbf{r}} \right] = \sum_{\mathbf{Q}} [i(\mathbf{k} + \mathbf{Q}) \cdot C_{n'k}(\mathbf{Q})] e^{i(\mathbf{k}+\mathbf{Q}) \cdot \mathbf{r}}, \]

which is efficiently evaluated using an FFT. Note that in the case where \( \mathbf{k} \) does not belong to the IBZ, a pseudo-wave function transformation has to be performed first according to Eq. (11).

Table 2 shows the timing results for the direct GPU porting. MPI initialization and distribution of the data was only performed at step 1 and there is no MPI communication throughout steps 2 - 11. As a result, the timing for the 1-CPU and 8-CPUs cases are expected to be the same. However, as shown in Table 2, the 8-CPUs case shows a 2-3\( \times \) slower performance because the different cores within 1-node compete for CPU memory bandwidth. In contrast, each GPU has its own dedicated memory, so 8-GPUs have the same timing as 1-GPU (results not shown). In reality, the RPA calculations will be executed on multiple nodes. As a result, the 8-GPUs / 8-CPUs comparison is more relevant.
Table 2: The timing (in units of seconds) for 1-CPU, 8-CPUs, 8-GPUs, as well as the 8-GPUs vs. 8-CPUs speed up (last column) for steps 2 - 11 in Table 1 for the test system N$_2$/Ru(0001) surface, modeled with 4 layers of Ru in a $\sqrt{3} \times \sqrt{3}$ unit cell. The timing information comes from a summation of 1 $k$-point (per core), 5 occupied and 1486 unoccupied bands with an energy cutoff of 150 eV. The bottom of the table summarizes the total timing for both the optical limit ($q \to 0$), and the $q \neq 0$ calculations. Note that in order to measure the GPU time for each step `cudaDeviceSynchronize` was used, while the total timing was obtained in the asynchronous mode. As a result, the “Total” time is smaller than the sum of the individual times. The CPU is an Intel Xeon X5650 and the GPU model is the “C 2075”.

| No. | Function  | 1-CPU (seconds) | 8-CPUs (seconds) | 8-GPUs$^{de}$ (seconds) | Speed up |
|-----|-----------|-----------------|-----------------|-------------------------|----------|
| 2)  | read_coef | -               | -               | 2.6                     | -        |
| 3)  | get_wfs   | 26.7            | 47.1            | 4.0                     | 11.6×    |
| 4)  | transform_wfs | 3.0            | 8.3             | 0.6                     | 12.7×    |
| 5)  | fft       | 18.9            | 30.1            | 2.7                     | 11.1×    |
| 6)  | mapG      | 0.2             | 0.4             | 0.2                     | 2.2×     |
| 7)  | paw_P_ai  | 6.0             | 7.3             | 2.6                     | 2.8×     |
| 8)  | paw_P_ap  | 4.1             | 9.9             | 6.5                     | 1.5×     |
| 9)  | paw_add   | 91.4            | 239.1           | 42.8                    | 5.6×     |
| 10) | optical_limit | 197.9          | 267.8           | 50.9                    | 5.3×     |
| 11) | zher      | 552.0           | 1193.2          | 89.6                    | 13.3×    |
|     | Total, $q \to 0$ | 911.3          | 1816.7          | 188.7                   | 9.6×     |
|     | Total, $q \neq 0$ | 665.9          | 1545.7          | 123.2                   | 12.5×    |

$ ^* $1-CPU calculates 1 $k$-point while 8-CPUs/8-GPUs calculate 8 $k$-points.

than the 1-GPU / 1-CPU comparison. Also note that the CPU results were obtained with the number of MKL threads set to 1. This is because threading is not implemented for ZHER in our current MKL library (version 10.3), and we use MPI to parallelize over cores.

According to Table 2 the average speed up for the pseudo-wave function portion (steps 3, 4, 5, 6, 10, 11) is around 10×, while for the PAW part (steps 7 - 9) it is around 4×. The most timing consuming part, step 11, which takes more than 60% of total simulation time, gains a speed up of only 11.4×. This is because the CUBLAS ZHER routine, which performs an outer product of a vector with length $n$ and adds that to a matrix of size $(n,n)$, is a memory bound operation. Such an argument applies to the other CUBLAS routines as well. The extremely poor performance for the PAW portion arises because the PAW contribution to the response function from each atom has a matrix size which is typically 5-50 (number of projector functions per atom) which is too small. In this case, the driver overhead for the cuda kernels exceeds the execution time.

3.2. Enhancing the GPU implementation

The GPU timing in Table 2 is dominated by steps 9 - 11, which use double-complex CUBLAS (GEMV and ZHER routine) and the CUFFT library. Both GEMV
Table 3: The timing (in units of seconds) and speed up (with respect to the \( Nu = 1 \) time) for the “multi-\( u \)” approach, with \( Nu = 1, Nu = 50 \) and \( Nu = 250 \) for the same steps and test system presented in Table 2. 8-GPUs are used throughout.

| No. | Function | \( Nu = 1 \) | \( Nu = 50 \) | \( Nu = 250 \) |
|-----|----------|--------------|--------------|--------------|
|     |          | seconds      | speed up     | seconds      | speed up     |
| 2)  | read coef| 2.5          | 1.0\( \times \) | 3.1          | 0.8\( \times \) |
| 3)  | get wfs  | 3.8          | 1.5\( \times \) | 2.6          | 1.4\( \times \) |
| 4)  | transform wfs | 0.6          | 1.6\( \times \) | 0.4          | 1.6\( \times \) |
| 5)  | fft      | 2.7          | 1.3\( \times \) | 2.0          | 1.3\( \times \) |
| 6)  | mapG     | 0.2          | 17.9\( \times \) | 0.007        | 29.6\( \times \) |
| 7)  | pawP\( u \) | 2.6          | 35.2\( \times \) | 0.03         | 89.5\( \times \) |
| 8)  | pawP\( n \) | 6.6          | 27.7\( \times \) | 0.1          | 43.2\( \times \) |
| 9)  | pawadd   | 42.8         | 18.4\( \times \) | 1.5          | 27.8\( \times \) |
| 10) | optical limit | 50.7         | 8.1\( \times \) | 5.6          | 9.1\( \times \) |
| 11) | zherk    | 89.6         | 12.9\( \times \) | 5.4          | 16.6\( \times \) |
|     | Total, \( q \to 0 \) | 188.5         | 9.0\( \times \) | 18.1         | 10.4\( \times \) |
|     | Total, \( q \neq 0 \) | 123.0         | 8.5\( \times \) | 12.2         | 10.0\( \times \) |

and \( \text{zher} \) are memory bound routines. Since the \( n(G) \) in Eq. (17) for each loop is completely independent from the other loops, we can group different \( n(G) \) together such that

\[
\chi_{GG'}^0(i\omega) = \sum_{k,u \subset n'} A(u,i\omega)n(u,G)n^*(u,G'),
\]

where \( u \) is a subset of index \( n' \), and \( n(u,G) \) is a matrix, with each column representing a vector \( n(G) \) at a particular \( n' \). We call this the “multi-\( u \)” approach.\(^1\) As a result, a BLAS level 2 \( \text{zher} \) problem is transformed into a BLAS level 3 \( \text{zherk} \) problem.

Table 3 (row 11) shows the timing and speed up of \( \text{zherk} \) compared to \( \text{zher} \) with different numbers of \( u \) (\( Nu \)), for \( n(G) = 1587 \). The speed up is 12.9\( \times \) with \( Nu = 50 \) and 16.6\( \times \) with \( Nu = 250 \). This improvement means the other steps such as 9 and 10 are now the performance bottlenecks. Thus we applied a similar idea to all the other steps. For simplicity, we keep the size of \( u \) uniform across the code.

As shown in Table 3, the largest speed up comes from the PAW portion. One subtlety with this code is that the number of projector functions is different for the different atomic species. In the CPU code, one has to loop over atoms and perform the operations sequentially for each atom, while in the GPU code the

\(^1\)A similar approach is used in the GPU implementation of the OCTOPUS package\(^{30}\), where the Kohn-Sham eigenstates are grouped together for time propagation of the Schrödinger equation.
loop over atoms, as well as the loop over projector functions (and bands when using the “multi-u” approach) can be eliminated by using thread parallelization. Each thread corresponds to an unique atom, projector function and band index. Since the operations on each atom are still linear algebra, the above algorithm is similar to batched CUBLAS function calls, although the sizes of the matrices are different within the batch. Since such a “flexible” CUBLAS batch is not yet available, we implemented our own customized CUDA kernel and achieved a significant improvement. In addition to the PAW and zherk portions, step 10 also has a non-trivial speed up of 9.1×, which results from a combination of batched cufft and our own kernels based on the “multi-u” approach.

The advantages of grouping small amount of independent data include that the number of kernel launches is reduced, reducing the effect of kernel launch overhead. Also cudaMemcpy is executed with larger amount of data per copy. This transforms the memory bound problem into a computation/latency bound problem.

3.3. Enhancing the CPU implementation

The above techniques for transforming memory bound problems into computational/latency bound problems can also be applied to the CPU implementation. In particular, a transformation from zher to zherk is straightforward. However, the thread parallelization over atoms in the PAW part is not possible for the CPU implementation. For the optical limit portion, the thread parallelization over bands and batched FFTW calls are non-trivial and are expected not to gain much performance in the CPU implementation. As a result, we only implemented zherk on the CPU. Given the same vector/matrix used in Table 2 and 3, the zherk versus zher speed up is 6.6× and 7.1× for Nu = 50 and 250, respectively.

3.4. Final GPU/CPU Performance Improvement

After enhancing both the CPU and GPU implementation using the “multi-u” approach, we summarize the 8-GPUs/8-Cpus speed up in Table 4 as a function of Nu. The Nu = 1 column is the same as the last column in Table 2 which corresponds to the “direct GPU porting” without employing the “multi-u” approach. As Nu increases, steps 3 - 5 have only slight changes and fluctuations. The major speed up as a function of Nu comes from the PAW part, which enables simultaneous thread parallelization over atoms, projector functions and bands. The speed up of CUBLAS vs. BLAS ZHERK increases from 13.3× up to 26.9× at Nu = 250. The matrix we used here (dimension of 1587×1587) is still not large enough to achieve the peak performance, which is around 36×. The final speed up (in asynchronous mode) is 30.6×/39.6× for optical/non-optical limit for the test system of N2/Ru(0001) surface.

4. Performance across different systems

The goal of the GPU port is not merely to improve performance, but to be able to address scientific problems with RPA. In this section we examine
Table 4: Final 8-GPUs vs 8-CPUs speed up after enhancing both the CPU and GPU implementation using the “multi-u” approach, with $Nu = 1, 2, 5, 10, 50, 150$ and $250$. The same steps and test system are used as in Table 2.

| No. | Function  | $Nu = 1$ | 2   | 5   | 10  | 50  | 150 | 250 |
|-----|-----------|----------|-----|-----|-----|-----|-----|-----|
| 3)  | get_wfs   | 11.6×    | 15.3×| 17.6×| 15.2×| 14.2×| 15.4×| 16.0×|
| 4)  | transform_wfs | 12.7× | 14.8×| 18.8×| 13.4×| 14.3×| 14.8×| 15.4×|
| 5)  | fft       | 11.1×    | 12.6×| 14.8×| 12.5×| 12.1×| 12.4×| 12.3×|
| 6)  | mapG      | 2.2×     | 4.4× | 10.9×| 14.0×| 33.8×| 47.5×| 53.4×|
| 7)  | paw_Pai   | 2.8×     | 5.0× | 12.1×| 23.4×| 94.0×| 195.9×| 242.2×|
| 8)  | paw_Pap   | 1.5×     | 2.6× | 7.0× | 10.5×| 36.2×| 51.5×| 56.7×|
| 9)  | paw_add   | 5.6×     | 13.0×| 30.7×| 48.6×| 82.1×| 114.8×| 136.2×|
| 10) | optical_limit | 5.3× | 4.7× | 9.5× | 11.2×| 17.8×| 19.9×| 19.2×|
| 11) | zherk     | 13.3×    | 13.0×| 13.2×| 11.8×| 22.9×| 26.2×| 26.9×|

Total, $q \to 0$ | 9.6× | 11.2×| 15.8×| 16.8×| 26.4×| 29.9×| 30.6×|
Total, $q \neq 0$ | 12.5×| 13.6×| 18.8×| 20.9×| 31.8×| 37.8×| 39.6×|

the performance of the GPU implementation across three types of systems: molecules, bulk solids and molecules adsorbed on surfaces.

Table 5 summarizes the selected systems, their performance improvement and the time required to complete the RPA calculation with a response function energy cutoff 150 eV. This cutoff is not high enough for a fully-converged result (250 eV is the minimum energy cut off to get converged results up to 50 meV) but this does not affect the conclusions here, since the speed up is in principle more favorable with larger energy cutoff, which corresponds to a larger number of plane waves and thus larger matrices. Also, the results are useful for comparison between similar systems or extrapolation of the results to different numbers of $k$-points used in the simulation. The number of bands used is equal to the number of plane waves for all the RPA calculations. Since a full CPU calculation is time consuming and unnecessary, the speed up presented in the table is obtained by performing some of the identical loops in the response function summation, while the time $t_{gpu}$ is obtained by completing the entire RPA calculation on 8-GPUs. The multi-u approach with $Nu = 100$ is used for both the CPU and GPU calculations.

$O_2$ is selected as a representative molecule. Although the number of plane waves increases linearly with the volume of the cell, it is reported that the RPA correlation energy converges rather fast with respect to the vacuum size used in the cell [7]. Using a (7Å, 7Å, 8.3Å) simulation cell, the RPA calculation of $O_2$ can be finished in 41 seconds on 8-GPUs. Similar timing applies to other small molecules such as $N_2$ and CO.

For bulk systems, a simple metal oxide $Li_2O$ and a transition metal oxide $MoO_3$ are selected. The unit cell for $MoO_3$ consists of 16 atoms and is larger
Table 5: The 8-GPUs/8-CPU speed up (column “Speed up”, for $q \neq 0$) as well as the time required to complete the entire RPA calculation with a response function cutoff of 150 eV (column $t_{gpu}$) using 8-GPUs for different systems (column “System”). For each system, the phase (column “Phase”), the number of atoms (column $N_a$) and the number of electrons (column $N_e$) in the unit cell, whether it is spin polarized (column “Spin”) and the number of $k$-points sampled for the BZ are specified. “sec” and “h” stand for seconds and hours, respectively.

| System         | Phase | $N_a$ | $N_e$ | Spin  | $k$-points | Improvement | $t_{gpu}$ |
|----------------|-------|-------|-------|--------|------------|-------------|-----------|
| O$_2$ gas      | gas   | 2     | 12    | True   | 1          | 11.3x       | 41 sec    |
| Li$_2$O bulk   | bulk  | 3     | 8     | False  | $4 \times 4 \times 4$ | 10.5x       | 63 sec    |
| MoO$_3$ bulk   | bulk  | 16    | 96    | False  | $4 \times 2 \times 4$ | 35.3x       | 1.0 h     |
| N$_2$/Ru(0001) surface | 14 | 202  | False | $4 \times 4 \times 1$ | 36.1x       | 1.4 h     |
| CO/Ni(111) surface | 22 | 210  | True  | $4 \times 4 \times 1$ | 37.0x       | 5.5 h     |

than that for Li$_2$O. The speed up increases from 10.5x to 35.3x due to the larger unit cell (because a larger number of plane waves are used). The total GPU time changes dramatically from 63 seconds for Li$_2$O to 1 hour for MoO$_3$ due to the theoretical $O(N^4)$ scaling for RPA calculations. The speed up and GPU timing on these bulk metal oxides are encouraging considering that we have spent over 1.5 million computing hours for calculating the formation energies of 23 metal oxides [37].

For surface systems, we selected two representative examples: N$_2$ adsorbed on Ru(0001) and CO on Ni(111) surfaces. The Ru(0001) surface is modeled with 4 layers having a $\sqrt{3} \times \sqrt{3}$ unit cell and the Ni(111) surface with 5 layers and a 2 $\times$ 2 unit cell. The latter unit cell and number of layers used are reported to converge the DFT chemisorption energies well [38]. The vacuum region is set to be 15 Å for both surfaces. Semi-core ($4s$ and $4p$) states are included in the Ru(0001) PAW potential. As a result, the number of electrons is similar for the Ru(0001) and Ni(111) surfaces, although the latter has a larger number of atoms included in the unit cell. A spin polarized calculation is employed for the Ni(111) case. The performance improvements for both systems are slightly better than for bulk MoO$_3$. It suggests that the speed up is approaching, although not quite achieving, the maximum possible, without fine-tuning of $N_u$ on a per-system basis. The final GPU time is 1.4 and 5.5 hour for N$_2$/Ru(0001) and CO/Ni(111), respectively. The longer time for CO/Ni(111) is because it is a spin polarized calculation with a larger 2 $\times$ 2 unit cell, which results in an increased number of bands included in the response function summation.

5. Conclusion and Outlook

We have ported the non-interacting density response function onto the GPU architecture. By grouping independent charge density matrices, we transformed
the problem from being memory bound into being computation/latency bound.
We call this the “multi-u” approach, where the number of \( u \) is the number of
independent vectors/matrices grouped together. The number of \( u \) is flexible and
constrained only by available memory. We enhanced both the CPU and GPU
implementations with the “multi-u” approach. The RPA calculations remain on
the GPU (no “thunking”). The size of the code is roughly 6000 lines of PYTHON
and 1000 lines of C/CUDA (many GPAW functions are re-used and not counted
here\(^2\)). The RPA correlation energy calculation performance improvement (8-
GPUs vs 8-CPPUs) is around 10\( \times \) for very small systems, and 40\( \times \) for standard
bulk systems and surfaces. With this improvement, an RPA calculation of CO
adsorbed on Ni(111) surface using 5 layers and \( 2 \times 2 \) unit cell, sampled with
16 \( k \)-points, can be finished in 5.5 hours using 8 GPUs. Such a speed makes it
promising to employ non-self-consistent RPA for routine surface chemistry simu-
lations, although it should be noted that the \( O(N^4) \) scaling for RPA calculations
have not changed by porting to GPU. Furthermore, since the non-interacting
response function is one of the most important and time consuming ingredients
for many beyond-DFT calculations such as TDDFT, GW, Bethe-Salpeter\(^3\),
we expect similar performance improvements in these beyond-DFT calculations
using the “multi-u” approach.

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\(^2\)The RPA GPU implementation is available for download at the GPAW svn repository
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