



Universität Hamburg

DER FORSCHUNG | DER LEHRE | DER BILDUNG

Bachelorthesis

Optimization of the Quantum Espresso Density Functional Theory Code for parallel execution on the PHYSnet-Cluster

vorgelegt von

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Kurzzusammenfassung

Abstract

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Contents

Danksagung

Motivation

Conventions

Throughout the text of this thesis scalars are written in italic s , vectors in bold italic \mathbf{v} and matrices in bold \mathbf{M} fonts. The summation/multiplication over nearest neighbour sites i and j is $\langle ij \rangle$ as a subscript to \sum/\prod . Furthermore, Hartree atomic units are used in general and only in selected instances it is deviated from this: $\hbar = m_e = e = 4\pi/\varepsilon_0 = 1$.

I Many-body physics

I.1 The electronic structure problem

In solid state physics, the Hamiltonian describing the interacting nuclei and electrons in the solid is well known, as all interactions except Coulomb interaction can safely be ignored at the mass and energy scales at which the electrons and nuclei reside. This very general problem consisting of both the electronic and nuclei degrees of freedom can be simplified in a first step by employing the Born-Oppenheimer approximation. The approximation assumes the nuclei to be fixed point charges which create a potential for the N interacting electrons, so that the electronic part can be solved independently using the nuclei positions \mathbf{R}_α as a parameter.

This problem is described by the time-independent Schrödinger equation

$$\hat{H}\Psi(\mathbf{r}_1, \dots, \mathbf{r}_N) = E\Psi(\mathbf{r}_1, \dots, \mathbf{r}_N) \quad (\text{I.1})$$

with the Hamiltonian in first quantization (i running over the electrons, α, β over the nuclei)

$$\hat{H} = \hat{T}_e + \hat{U}_{e-e} + \hat{V}_{n-e} + \hat{W}_{n-n} \quad (\text{I.2})$$

$$= -\sum_i \frac{1}{2} \nabla_i^2 + \frac{1}{2} \sum_{i \neq j} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} - \sum_i \sum_\alpha \frac{Z_\alpha}{|\mathbf{r}_i - \mathbf{R}_\alpha|} + \frac{1}{2} \sum_{\alpha \beta} \frac{Z_\alpha Z_\beta}{R_{\alpha\beta}} \quad (\text{I.3})$$

where:

- \hat{T}_e is the kinetic energy of the electrons
- \hat{U}_{e-e} is the Coulomb interaction between the electrons and
- \hat{V}_{n-e} is the potential energy of the electrons in the field of the nuclei
- \hat{W}_{n-n} is the Coulomb interaction between the nuclei

The terms \hat{V}_{n-e} and \hat{W}_{n-n} can then be combined into an external potential V for the interacting electrons, so that the Hamiltonian reads

$$\hat{H} = \hat{T} + \hat{U} + \hat{V} \quad (\text{I.4})$$

This Hamiltonian will be used in the further development of the underlying theory for this thesis.

I.2 Density Functional Theory

A direct solution to the electronic structure problem, this meaning obtaining the ground-state many-body wave function $\Psi(\mathbf{r}_1, \dots, \mathbf{r}_N)$ for a given potential is analytically impossible even

for a small number of electrons compared to the number of electrons in a macroscopic crystal. As such, the need for good approximations to obtain results for real world systems is high. One particularly successful approach is **Density Functional Theory (DFT)**. In the following section, the theoretical framework of **DFT** will be developed, the outline of which can be found in any literature on solid state physics [1].

I.2.1 Hohenberg-Kohn theorems

The starting for DFT is the exact reformulation of the outlined electronic structure problem by Hohenberg and Kohn [2]. This reformulation uses the ground state density of the electronic system $n_o(r)$ as the basic variable. To achieve this, Hohenberg and Kohn [2] formulated two theorems, which demonstrate that the ground state properties of an electronic system can be described using the ground state density (the proof of those theorems is omitted here, but can be found in the original paper [2] or any publication on **DFT** [1]):

- I The external potential (and via the Schrödinger equation also the ground state wave function and the ground state energy) is a unique functional of the ground state density (except for an additive constant).
- II The ground state energy minimizes the energy functional,

$$E[n(\mathbf{r})] > E_0 \quad \forall \quad n(\mathbf{r}) \neq n_0(\mathbf{r})$$

The proof of those theorems show the existence of the energy functional $E[n(\mathbf{r})]$, but a concrete expression for it cannot be given. As the ground state wave function is a functional of the ground state density, a formal definition of the energy functional can be written as

$$\begin{aligned} E[n(\mathbf{r})] &= \langle \Psi | \hat{H} | \Psi \rangle \\ &= \langle \Psi | \hat{T} + \hat{U} + \hat{V} | \Psi \rangle \\ &= \langle \Psi | \hat{T} + \hat{U} | \Psi \rangle + \int d\mathbf{r}' \Psi^*(\mathbf{r}') V(\mathbf{r}') \Psi(\mathbf{r}') \end{aligned}$$

Defining the universal functional $F[n(\mathbf{r})] = \langle \Psi | T + U | \Psi \rangle$, which is material independent and writing $n(\mathbf{r}') = \Psi^*(\mathbf{r}') \Psi(\mathbf{r}')$, the energy functional becomes

$$E[n(\mathbf{r})] = F[n(\mathbf{r})] + \int d\mathbf{r}' V(\mathbf{r}') n(\mathbf{r}') \quad (\text{I.5})$$

This is just a formal definition, as all the formerly mentioned complication of the Hamiltonian **I.4** now lie in the functional $F[n(\mathbf{r})]$. With a known or well approximated universal functional $F[n(\mathbf{r})]$, the Hohenberg-Kohn theorems provide a great simplification for finding the ground state properties of a solid state system, as the problem is now only a variational problem with 3 spatial coordinates instead of $3N$ coordinates when trying to solve the full Hamiltonian.

I.2.2 Kohn-Sham equations

One way of approximating the functional $F[n]$ was given by Kohn and Sham [3]. The idea is to use a non-interacting auxiliary system of electrons

$$H_0 = \sum_i^{N_e} \frac{p_i^2}{2m} + v_{KS}(\mathbf{r}_i) \quad (\text{I.6})$$

With a correction potential v_{KS} such that the ground state charge density for the auxiliary and the interacting system are the same. This introduces a new set of orthonormal wave functions, the solutions to the non-interacting problem Ψ_i . The kinetic energy of such a non-interacting problem can be easily calculated as sum over all electrons:

$$T_S[n(\mathbf{r})] = -\frac{1}{2} \sum_{i=1}^{N_e} \int d^3r \Psi_i^*(\mathbf{r}) \Delta \Psi_i(\mathbf{r}) \quad (\text{I.7})$$

With the help of this system and the classical electrostatic energy

$$E_H[n(\mathbf{r})] = \frac{1}{2} \int \int d^3r_1 d^3r_2 \frac{n(\mathbf{r}_1)n(\mathbf{r}_2)}{|\mathbf{r}_1 - \mathbf{r}_2|} \quad (\text{I.8})$$

an ansatz for the universal functional can be written as

$$F[n(\mathbf{r})] = T_S[n(\mathbf{r})] + E_H[n(\mathbf{r})] + E_{XC}[n(\mathbf{r})] \quad (\text{I.9})$$

where now $E_{XC}[n(\mathbf{r})]$ is a functional of the density accounting for all exchange and correlation effects not present in the non-interacting electron system. The success of **DFT** lies in the fact that E_{XC} only contributes only a small part of the total energy and can be surprisingly well approximated.

Using that form of $F[n(\mathbf{r})]$, from the variational problem (with a Lagrange parameter introduced to ensure the orthonormality of the states Ψ_i)

$$\delta \left(E[n(\mathbf{r})] - \sum_j \lambda_j \left[\int d^3r |\Psi_j|^2 - 1 \right] \right) = 0 \quad (\text{I.10})$$

single particle, Schrödinger-like equations can be derived:

$$\left(-\frac{1}{2}\Delta + \frac{1}{2} \int d^3r' \frac{n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + \frac{\delta E_{XC}}{\delta n(\mathbf{r})} + V \right) \Psi_i(\mathbf{r}) = \lambda_i \Psi_i(\mathbf{r}) \quad (\text{I.11})$$

Schrödinger-like in this context means, that with the identification

$$v_{KS} = V_H + V_{XC} + V = \frac{1}{2} \int d^3r' \frac{n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + \frac{\delta E_{XC}}{\delta n(\mathbf{r})} + V \quad (\text{I.12})$$

eq. **I.11** become the **KS** equations:

$$\left(-\frac{1}{2}\Delta + v_{KS} \right) \Psi_i(\mathbf{r}) = \epsilon_i \Psi_i(\mathbf{r}) \quad (\text{I.13})$$

Importantly, the potential v_{KS} depends on the solutions $\Psi_i(\mathbf{r})$, as the Hartree potential V_H and the XC potential V_{XC} include the density $n(\mathbf{r})$, so the problem becomes a self-consistency problem.

density of KS
states

I.2.3 Pseudopotentials and basis set

In order to represent the states and operators in eq. I.11, a basis set has to be chosen. A natural choice, at least for delocalized (higher energy) orbitals is the discrete set of plane waves $|\mathbf{G}\rangle$

$$\langle \mathbf{r} | \mathbf{k} + \mathbf{G} \rangle = \frac{1}{\sqrt{V}} e^{i(\mathbf{k} + \mathbf{G})\mathbf{r}} \quad (\text{I.14})$$

where \mathbf{G} is a reciprocal lattice vector. One problem of this approach lies in the fact that the lower energy core electrons are in contrast very localized around the cores and as such need a lot of basis functions. Because the higher orbitals have to be orthogonal to the lower orbitals, they must also have features on the length scale of the core electrons, which means more basis functions are needed to describe them accurately.

Importantly, the core electrons don't contribute much to the electronic structure problem, so an approach to make the calculations more economically is to introduce an effective screened potential which describes the nuclear potential as well as the states close to the nucleus. These potentials are called **Pseudopotentials (PP)** and

not finished

I.3 Density Functional Perturbation Theory

hate thinking
about it

II Computational Basics

II.1 Parallel computing

The following section will give an overview of the technical aspects of running computer code (such as QUANTUM ESPRESSO) on massively parallel computing environments (such as the PHYSnet compute cluster). The information presented can be found in any textbook on parallel or high-performance computing [4].

functional/data
parallelism?

II.1.1 On scalability

In scientific computing, one can identify two distinct reasons to distribute workloads to multiple processors:

- The execution time on a single core is not sufficient. The definition of sufficient is dependent on the specific task and can range from “over lunch” to “multiple weeks”
- The memory requirements grow outside the capabilities of a single core

In order to judge how well a task can be parallelized, usually some sort of scalability metric is employed, for example:

- How fast can a problem be solved with N processors instead of one?
- What kind of bigger problem (finer resolution, more particles, etc.) can be solved with N processors?
- How much of the resources is used for solving the problem?

The speedup by using N workers to solve a problem instead of one is defined as $S = \frac{T_1}{T_N}$, where T_1 is the execution time on a single processor and T_N is the execution time on N processors. In the ideal case, where all the work can be perfectly distributed among the processors, all processors need the same time for their respective workloads and don't have to wait for others to finish their workload to continue, the execution time on N processors would be $\frac{T_1}{N}$, so the speedup would be $S = \frac{T_1}{\frac{T_1}{N}} = N$.

In reality, there are many factors either limiting or in some cases supporting parallel code scalability. Limiting factors include:

- *Algorithmic limitations*: when parts of a calculation are mutually dependent on each other, the calculation cannot be fully parallelized
- *Bottlenecks*: in any computer system exist resources which are shared between processor cores with limitations on parallel access. This serializes the execution by requiring cores to wait for others to complete the task which uses the shared resources in question

- *Startup Overhead*: introducing parallelization into a program necessarily introduces an overhead, e.g. for distributing data across all the processors
- *Communication*: often solving a problem requires communication between different cores (e.g. exchange of interim results after a step of the calculation). Communication can be implemented very effectively, but can still introduce a big prize in computation time

On the other hand, faster parallel code execution can come from:

- *Better caching*: when the data the program is working with is distributed among processors (assuming constant problem size), it may enable the data to be stored in faster cache memory. Modern computers typically have three layers of cache memory, with level 1 cache being the smallest and fastest and level 3 being the largest and slowest, so smaller data chunks per processor can lead to the data not being stored in main memory, but completely in cache or in a faster cache level

strong/weak scaling?

II.2 Quantum ESPRESSO

QUANTUM ESPRESSO (opEn-Source Package for Research in Electronic Structure, Simulation, and Optimization) [5, 6] is a collection of packages implementing (among others) the techniques described in sec. 1.2 and ?? to calculate electronic structure properties

what is needed for compilation?
also in next subsection!

II.2.1 Parallelization of Quantum ESPRESSO calculations

uff

III Parallelisation of self-consistent calculations of electronic-structure properties

III.1 First scaling tests

The first step in analysing the scaling of QUANTUM ESPRESSO is to perform a baseline scaling test without any optimisations applied. In Fig. III.1 and ?? two scaling tests on the earlier mentioned benchmarking systems Si and TaS2 are pictured. The tests are run using QUANTUM ESPRESSO 7.0, compiled using the Fortran and C compilers in OpenMPI 4.1.0, without any of compilation or runtime optimisation parameters mentioned in section II.2 used.

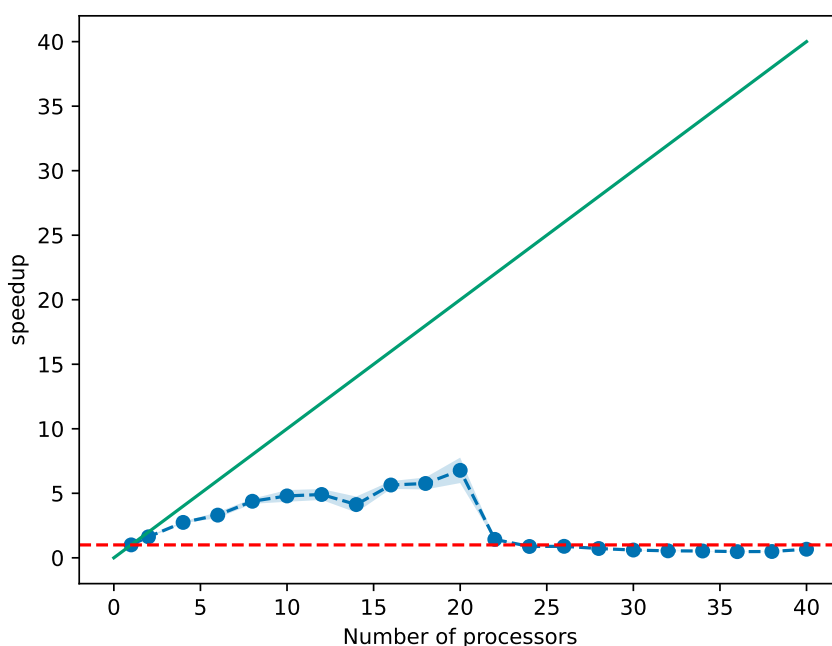


Figure III.1: Baseline scaling test on the Si benchmarking system QUANTUM ESPRESSO 7.0, OpenMPI 4.1.0, nk 1 and nd 1

Three different metrics of scalability are pictured in ??.

- runtime: absolute runtime (walltime) of the compute job

talk a bit about silicon there, then introduce TaS2

formulate differently with new figure structure!

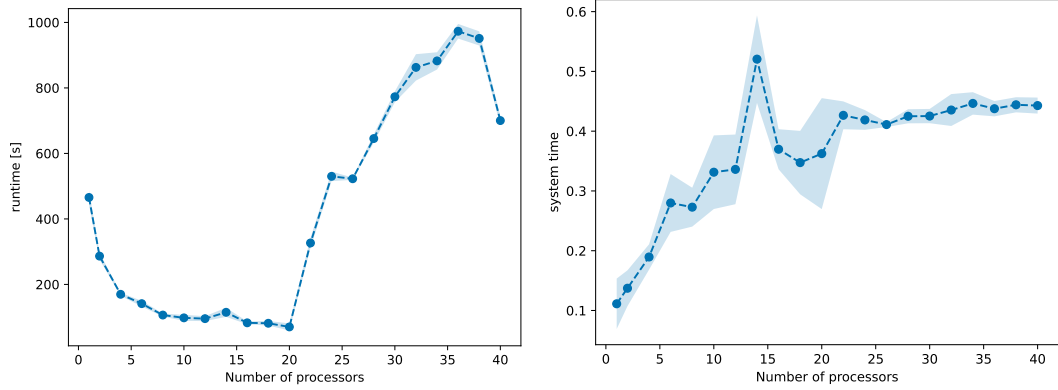


Figure III.2: Baseline scaling test on the Si benchmarking system QUANTUM ESPRESSO 7.0, OpenMPI 4.1.0, `nk 1` and `nd 1`

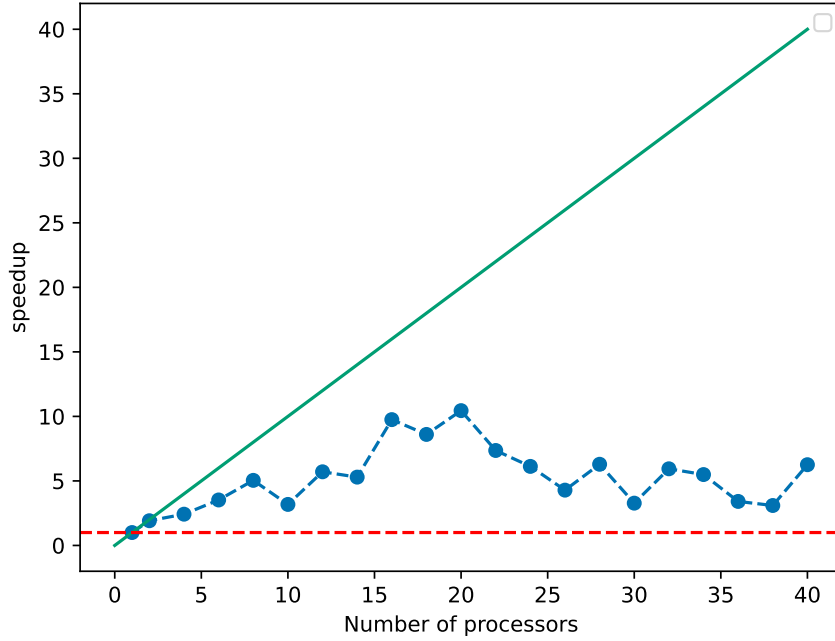


Figure III.3: Baseline scaling test on the TaS2 benchmarking system QUANTUM ESPRESSO 7.0, OpenMPI 4.1.0, `nk 1` and `nd 1`

- speedup: runtime divided by runtime of the job on a single core
- system time: percentage of wall time used by system tasks, e.g. writing to disk, etc. (calculated as $(\text{walltime} - \text{cputime}) / \text{walltime}$)

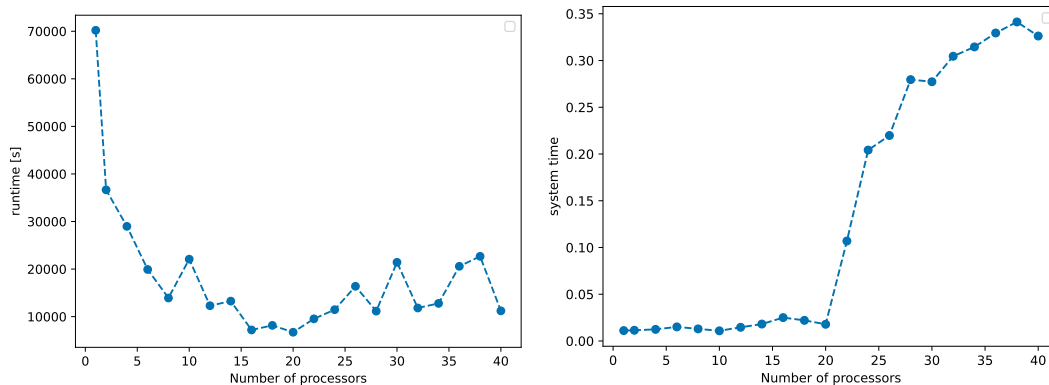


Figure III.4: Baseline scaling test on the TaS2 benchmarking system QANTUM ESPRESSO 7.0, OpenMPI 4.1.0, `nk 1` and `nd 1`

For further analysis mainly speedup will be used as a metric of scalability, because it lends itself to easy interpretation: optimal scalability is achieved when the speedup scales linearly with the number of processors (with a slope of one), as discussed in ch. II.1. This on the one hand necessarily implies good parallelization and a lower runtime for more processors used, but the other two parameters should also always be considered.

As an example, for a problem with a single core runtime of 600 s, a speedup of 100 would mean a runtime of 6 s, whereas a speedup of 200 would mean a runtime of 3 s. Even with optimal scaling, the 100 processors needed for the speedup of 200 could be considered wasted for just 3 s of saved time. On the other hand, for a problem with a single core runtime of 2400 h, the difference between a speedup of 100 (runtime 24 h) and 200 (runtime 12 h) is the difference between needing to wait a whole day for the calculation and being able to let the job run overnight and continue working in the morning, so using 100 more processors for that can be considered a good use of resources.

On a single node, both the Si and TaS2 calculations show good, but not perfect scaling behavior: the speedup does approximately scale linearly with the number of processors, but the slope is closer to $\frac{1}{2}$, than 1. Even though the scaling behavior is not perfect, there is just a small, almost constant amount of runtime used by system calls, this speaks for good parallelization. As discussed in sec. II.1, startup time is an unavoidable part of every parallel program, so a constant amount of time used not for calculations is expected, bad parallelization on the other hand shows itself by introducing waiting times between processors, which makes the waiting time in some way dependent on the number of processors.

When using more than one node, not only does the scaling get worse, the execution needs longer than on a single core for the Si system, with a marginally better performance for the TaS2 system. This is also seen in the plots of system time. The percentage of time used for tasks not directly related to calculations goes from a near constant value for under 20 processors to 50% of the execution time for the Si system and 35% for the TaS2 system.

These scaling tests pose now two questions to be answered:

- Is better scaling on a single node possible?

explanation of
wait time

- How can acceptable scaling over more than one node be achieved?

III.2 Testing different compilers and mathematical libraries

A first strategy for solving issues with parallelization is trying different compilers and mathematical libraries. In the PHYSnet cluster a variety of software packages is available. As discussed in sec. II.2, for the compilation of QUANTUM ESPRESSO

missing

For testing QUANTUM ESPRESSO will be compiled using the following software combinations:

- OpenMPI 4.1.0 and OpenBLAS
- OpenMPI 4.1.0 and Scalapack
- Intel OneAPI 2021.4 (includes Intel MPI, Fortran and C compilers as well as Intel MKL, a scalable mathematical library)

analysis

III.3 Using the parallelisation parameters of Quantum ESPRESSO

As detailed in section II.2, QUANTUM ESPRESSO offers ways to manage how the workload is distributed among the processors. In `pw.x` the default plane wave parallelization, k-point-parallelization and linear-algebra parallelization are implemented.

The benchmark pictured in III.6 is set up as follows: for a given number of processors N_p , the parameter N_k splits the N_p processors into N_k processors pools. As the number of processors in one pool has to be a whole number, only certain combinations of N_p and N_k are possible, for example $N_p = 32$ could be split into processor pools of size 2 with $N_k = 16$, size 8 with $N_k = 4$ or size 16 with $N_k = 2$. This leads to choosing the size of the processor pools as a variable, not the parameter `nk`. Fig. III.6 shows the scaling for poolsizes 2, 8 and 16 for QUANTUM ESPRESSO being compiled with OpenMPI/Scalapack and Intel oneAPI. This choice of pool sizes showcases the smallest pool size possibly (namely 2), as well as a bigger pool size with 16, that still gives rise to a few data points over the chosen range of processors.

Fig. III.6 shows that using k parallelization with a pool size of 2 significantly improves the scaling behavior, not only on one node, but especially over more than one node.

analysis

Another important conclusion to draw out of fig. III.6 is the impact of using Intels compiler instead of OpenMPI, as that factor alone speeds up the calculation by a factor of 2 over the whole range of processors.

The same scaling test is applied to the TaS2 system in fig. III.7, with the same list of pool sizes, but over a wider range of processors.

Remarkably, the scaling behavior is swapped in comparison to III.6, as the pool size 2 saturates fast and the bigger pool sizes show way better scaling behavior. Furthermore, there are instances of better than linear scaling, which according to QUANTUM ESPRESSO docs can be attributed to better caching of data.

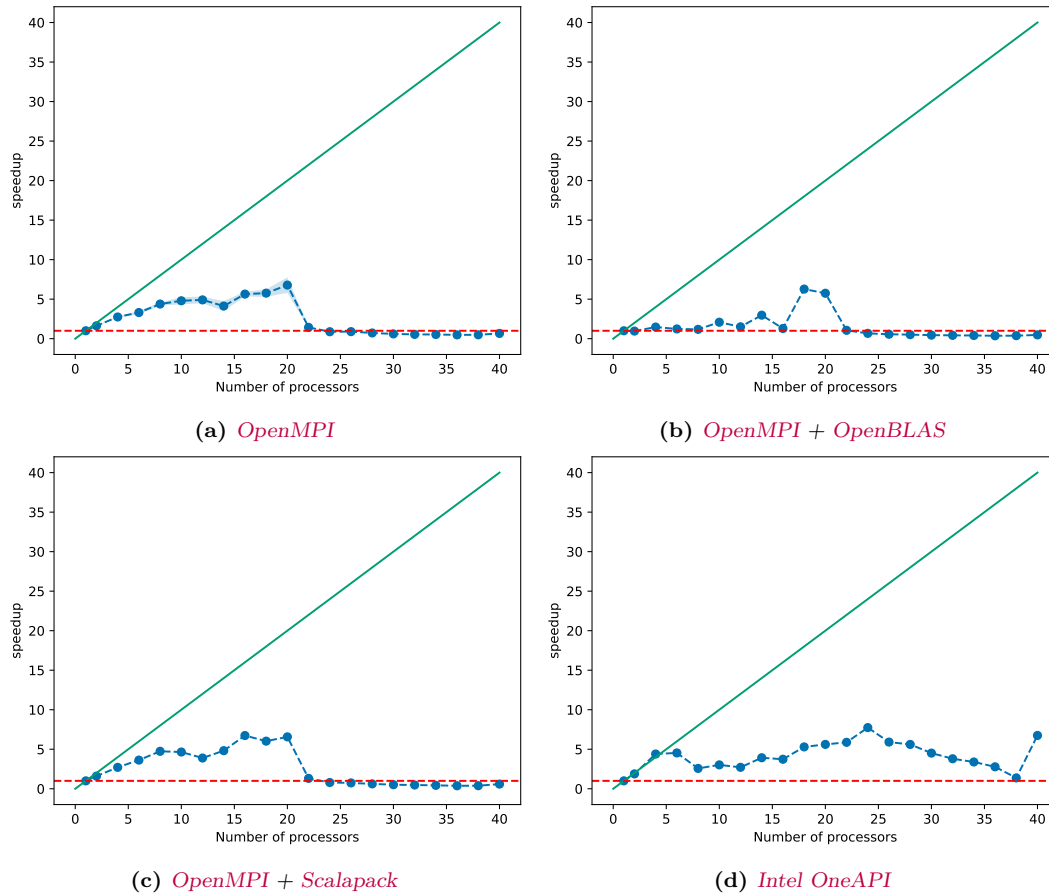


Figure III.5: Baseline scaling test with different combinations of compilers and mathematical libraries

It can also be instructive to look at the idle time for this benchmark to judge the quality of parallelization.

Fig. III.8 shows a distribution of idle times between 1 % and 4 % of the whole wall time, without any kind of systemic increase over any range of processors. This means the parallelization is as good as possible for these

missing

Linear Algebra

III.3.1 Comparison with calculations on the HLRN cluster

III.3.2 Conclusion: Parameters for optimal scaling

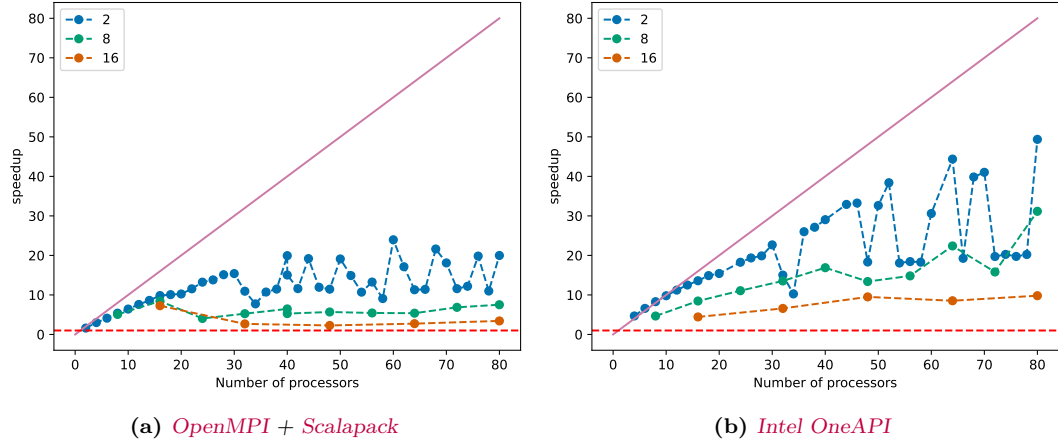


Figure III.6: Benchmark with k -point parallelization for the Si benchmarking system with 3 different sizes of processor pools

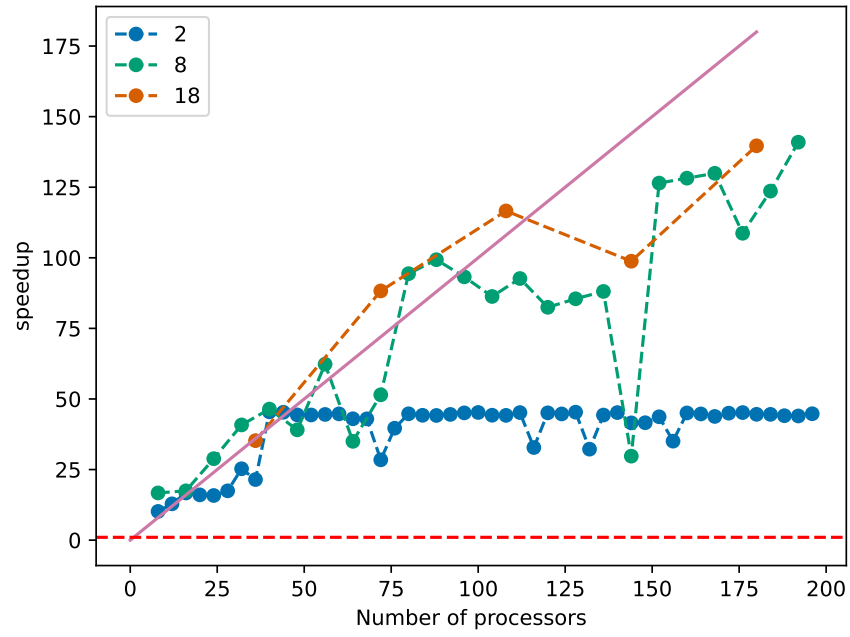


Figure III.7: Benchmark with k -point parallelization for the TaS2 benchmarking system

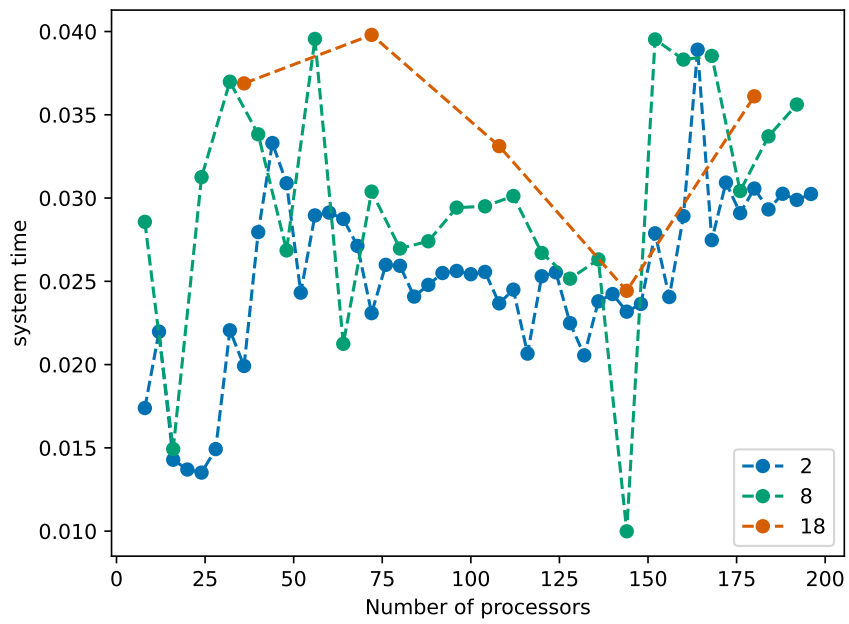


Figure III.8: Idle time for the k point parallelization benchmark for the TaS₂ system

IV Parallelization of DFPT calculations

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