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Mohammed Benaissa, Tarik Ouahrani, Keisuke Hatada, Kazuki Yoshikawa, Didier Sebilliau. Simple approach to more efficient density functional theory simulations. *Computational Condensed Matter*, 2025, 42, pp.e01012. <10.1016/j.cocom.2025.e01012>. <hal-04984658>

**HAL Id: hal-04984658**

**<https://hal.science/hal-04984658v1>**

Submitted on 3 Apr 2025

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# Simple Approach to More Efficient Density Functional Theory Simulations

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## Abstract:

In physics, chemistry and materials science, density functional theory (DFT) is a common method for studying the electronic structure of many-body systems. DFT can address complex materials with relatively low computational costs compared to other physics-based methods; however, it still requires considerable computational power for large-scale simulations. With the aim of reducing the computational footprint of DFT, we suggest the use of the data-efficient Bayesian algorithm to optimize the charge mixing parameters, which reduces the self-consistent field iterations necessary to reach convergence. We show that our algorithm can achieve faster convergence than the default parameters when the VASP code is used as a proof of concept, resulting in significant time savings in DFT simulations and therefore providing a systematic guide for more efficient DFT simulations. We propose adding this procedure to the well-known convergence test procedures, such as cutoff-energy and k-point convergence tests.

**Keywords:** Self-Consistent Field, Charge Mixing Parameters, Bayesian Optimization.

## 1. Introduction

Computational methods have brought about a major transformation in the realm of material analysis and design by providing a supplementary avenue for experimentation. These methods have enabled researchers to precisely regulate simulation conditions, a feat that is arduous under experimental conditions. In the past three decades, material science has been revolutionized by electronic structure methodologies, particularly density functional theory (DFT). This theory offers a relatively accurate and efficient means of predicting properties and designing novel materials by solving the Schrödinger equation for the system's electrons [1]. DFT-based ab-initio simulations have immense influence in fields such as chemistry, physics, biology, and material science at large [2], facilitating the study of complex systems [3] with hundreds and thousands of atoms (e.g., DNA [4], proteins [5], and even electronic devices [6]). High-throughput calculations enabled by DFT simulations have successfully predicted novel materials suitable for applications such as redox-flow batteries [7], catalysts [8], two-dimensional topological insulators [9], and photovoltaics [10]. These developments have contributed to the creation of extensive databases of computed properties freely available for the scientific community.

While DFT-based computational methods have been used as game changers in materials science research, it is essential to acknowledge their high energy consumption and work toward more efficient computing solutions. Possible solutions include using renewable energy sources to power high-performance computers; developing efficient algorithms and codes, such as linear scaling DFT [11]; and designing high-performance computing systems with low power requirements. Another popular approach is the integration of machine learning to accelerate DFT simulations [12,13]. Inexpensive machine learning (ML) techniques are employed to learn data structures from existing DFT simulations and map the properties to their respective descriptors [14]. Additionally, researchers can optimize their simulations and implement techniques to remove unnecessary calculations.

One challenge is to reduce the computational costs of DFT calculations by optimizing the parameters that control charge density mixing in *self-consistent* DFT calculations. Density mixing is considered a fundamental *self-consistent* field (SCF) method employed in electronic structure calculations that aims to iteratively converge to the Kohn–Sham ground-state density.

This is achieved by mixing previous densities until the final charge density equals the previous density to a certain threshold, ensuring a *self-consistent* solution. Specifically, given an initial input density and the resulting output density from the previous iteration, charge mixing methods could produce a more refined input density estimate for the subsequent *self-consistent* iteration (SCI). We focus on optimizing the number of self-consistent iterations in DFT calculations, which is a key factor in determining computational costs. This task is performed by identifying the optimal set of input parameters for charge mixing that leads to the lowest number of self-consistent iterations possible using an optimization algorithm.

Optimization methods can be broadly divided into two categories: derivative-based methods [15] and derivative-free methods [16]. Derivative-based optimization methods are a class of optimization algorithms that utilize the derivative information of the objective function to guide the search toward the optimal solution. These methods are particularly effective when the objective function is well-behaved and possesses continuous derivatives [17]. By exploiting the gradient or Hessian of the objective function, derivative-based algorithms can efficiently navigate the search space and converge rapidly to optimal solutions. Examples of derivative-based optimization methods include gradient descent, Newton's method, and quasi-Newton methods.

While derivative-based methods excel at optimizing well-behaved functions, their effectiveness diminishes when dealing with functions that are noisy, discontinuous, or lack derivatives altogether, which is the case here. In such scenarios, derivative-free optimization methods emerge as valuable alternatives [18]. Derivative-free optimization methods, also known as black-box optimization methods, are designed to optimize objective functions without relying on derivative information. These methods are particularly useful when the objective function is expensive to evaluate, making it impractical to compute its derivatives. Derivative-free algorithms employ various strategies to explore the search space and identify optimal solutions without explicit knowledge of the objective function's gradient or Hessian. Examples of derivative-free optimization methods include the Nelder–Mead simplex [19], Simulated annealing [20], Genetic algorithms [21], and Bayesian optimization (BO) [22]. This latter has been successfully applied in numerous fields, such as ML hyperparameter tuning to achieve optimal performance [23], engineering to design high-quality products [24], chemistry to develop new materials [25], robot learning to improve task performance [26], and even culinary arts [27].

BO is a highly effective method for optimizing complex black-box functions with minimal data requirements. The function can be evaluated for different input combinations, and the internal workings of the function can be understood by constructing surrogate models [28]. Bayesian optimization works by repeatedly creating surrogate models of the black-box function and strategically assessing the function at the most promising or informative input location on the basis of the information observed thus far. Hence, BO is a versatile and powerful tool that can be applied in various research domains. Due to the data-efficient character of BO, we applied it to optimize the charge mixing parameters for more efficient DFT calculations. We demonstrate the usefulness of this approach for different systems, insulating, semiconducting and metallic. This research provides a systematic approach for computationally more efficient DFT simulations for the scientific community. This practice can be performed alongside the regular cutoff and k-point convergence test procedures.

## 2. Computational Methods

### 2.1. DFT calculations

In this study, first-principles simulations based on the Kohn–Sham formalism of density functional theory (DFT) [29,30] were performed using the Vienna Ab initio Simulation Package (VASP) [31,32] on a number of extensively studied materials [all the citations]. The structural information of these materials is provided in the supplementary material (Table S.1). The Perdew–Burke–Ernzerhof functional (PBE) [33] of the generalized gradient approximation LDA [34] and GGA [35] [36] and R<sup>2</sup>SCAN [37–39] and the projector augmented wave method (PAW) [40], were utilized for the exchange–correlation functional and electron–ion interactions. The present simulation was conducted on primitive structures. However, we employ a supercell method for calculating the mechanical properties of the materials. These calculations use a plane-wave basis cutoff energy of 520 eV and the Monkhorst-Pack special k-point mesh for integrating the Brillouin zone, as indicated in Table S.2 from the supplementary materials (SM).

The conjugate gradient (CG) method, as implemented in VASP code [41] has been employed for all materials to optimize the crystal structure, satisfying the energy convergence

criterion of  $10^{-8}$  eV for self-consistent field (SCF) calculations and the Hellmann–Feynman (HF) forces convergence criterion of  $10^{-4}$  eV/Å.

## 2.2. Charge mixing

Charge mixing is the iterative process of updating the charge density in self-consistent field (SCF) calculations within density functional theory (DFT). Effective charge mixing strategies accelerate the convergence of electronic minimization by controlling oscillations in charge density updates, thus avoiding issues like charge sloshing that can stall or slow convergence. Key electronic structure codes, including VASP, employ various charge mixing schemes, notably Kerker mixing [42], Tchebycheff-inspired mixing [43], a modified Broyden method [44], and Pulay mixing [45]. In VASP, the choice of mixing scheme is controlled by the IMIX tag, with the following options:

- IMIX = 0: No mixing where no charge density updates are performed during the iteration.
- IMIX = 1: Kerker mixing, where the mixed density is given by [42]:

$$\rho_{\text{mix}}(G) = \rho_{\text{in}}(G) + A \frac{G^2}{G^2 + B^2} (\rho_{\text{out}}(G) - \rho_{\text{in}}(G)) \quad (1)$$

with  $A = \text{AMIX}$  and  $B = \text{BMIX}$ .

- IMIX = 2: Variant of Tchebycheff mixing, where the following second order equation of motion is used:

$$\ddot{\rho}_{\text{in}}(G) = 2 * A \frac{G^2}{G^2 + B^2} (\rho_{\text{out}}(G) - \rho_{\text{in}}(G)) - \mu \dot{\rho}_{\text{in}}(G) \quad (2)$$

with  $A = \text{AMIX}$ ,  $B = \text{BMIX}$ , and  $\mu = \text{AMIN}$ .

- IMIX = 4: Broyden's modified method [44] and Pulay-mixing method [45], depending on the weight factor WC. Broyden's method iteratively improves an approximate Jacobian matrix to optimize the solution, making it effective in systems where linearity is assumed in the residuals. Pulay mixing, on the other hand, combines input vectors linearly to minimize the residual, often yielding faster convergence for a wide range of materials.

The default setting uses a Pulay mixer initialized with an approximate charge-dielectric function based on Kerker's method [42]:

$$A \times \min\left(\frac{G^2}{G^2 + B^2}, A_{min}\right) \quad (3)$$

In the input setting of the VASP (INCAR), these variables are translated to  $A = AMIX$ ,  $B = BMIX$ , and  $A_{min} = AMIN$ .

### Additional Mixing Parameters in VASP

- **MIXPRE:** Determines the metric for preconditioning in the Broyden mixing scheme. Preconditioning can improve stability by applying an "Inverse Kerker" metric, which is particularly useful for challenging systems.
- **MAXMIX:** Specifies the maximum number of steps stored in the Broyden mixer, effectively controlling the rank of the approximated charge-dielectric matrix. The value of MAXMIX can be either positive or negative;  $MAXMIX < 0$ : The mixer is reset after each ionic step or if the electronic steps exceed the absolute value of MAXMIX.  $MAXMIX > 0$ : Storage in the mixer persists, and oldest vectors are smoothly removed from the history when storage is exceeded.
- **NELMDL:** Specifies the number of non-self-consistent electronic steps at the beginning of a calculation, allowing the electronic wavefunctions to relax before the charge density is fully updated.

## 2.2 Bayesian algorithm

The application of Bayesian optimization to charge mixing parameter optimization offers several advantages over traditional optimization approaches. Traditional methods often rely on gradient-based descent techniques, which can cause the algorithm to become trapped in local minima and fail to converge to the global minimum. In contrast, Bayesian optimization employs a probabilistic framework that allows it to explore the parameter space more efficiently and avoid local minima. The first step in implementing Bayesian optimization is to define the parameter space. This involves identifying the parameters that affect the DFT calculations and determining the range of values that each parameter can take. Once the parameter space is defined, a surrogate

model is constructed to approximate the function of the parameter space [46]. The surrogate model is a statistical model that maps input parameters to corresponding output values. In this work, a Gaussian process model was employed as the surrogate model. Gaussian processes are known for their flexibility and ability to handle noisy data, making them suitable for applications such as simulation parameter optimization where noise is often present.

The acquisition function plays a crucial role in guiding the search for optimal parameters. This approach balances the trade-off between exploration and exploitation, allowing the algorithm to efficiently explore the parameter space while also focusing on regions that are likely to contain optimal parameter values. For this task, the Tree of Parzen Estimators (TPE) acquisition function was chosen [47]. TPE is particularly well suited for mixed continuous and discrete inputs, making it a suitable choice for the parameters involved in DFT calculations. Using the defined acquisition function, the surrogate model was iteratively evaluated at the most promising parameters values. This process was continued until the desired number of iterations was reached. In this case, 30 iterations were found to be sufficient to achieve satisfactory results. Subsequently, the optimized parameters were rigorously validated by comparing the DFT calculation results obtained using these parameters to those obtained using the default parameters in the VASP program. A comparison of the results showed that the optimized parameters produced the same results in considerably less time, demonstrating the effectiveness of this approach.

We developed a Python script utilizing the Hyperopt library to optimize the VASP simulation parameters. The optimization process is driven by a black-box function that modifies the INCAR file to adjust several input parameters: IMIX, AMIX, BMIX, AMIN, NELMDL, WC, MIXPRE, and MAXMIX. The parameter ranges are as follows: IMIX can take values 1, 2, or 4. When IMIX=4, the additional parameters WC, MIXPRE, and MAXMIX are adjusted. AMIX, BMIX, and AMIN are specified as floating-point values between 0 and 2, while NELMDL is chosen from integer values between -5 and -1. WC is set as an integer between 0 and 1000, MIXPRE can be selected from 0, 1, or 2, and MAXMIX varies between -200 and 200.

The script explores different combinations of these parameters, performs density functional theory (DFT) calculations with VASP, and evaluates the number of self-consistent iterations (SCI) needed for convergence. The optimization process runs for 30 iterations, during

which a Gaussian process model updates the input-output mapping and guides the search for the next best parameter combination after each iteration. Based on our experience with the materials under study, 30 iterations are typically sufficient to converge on an optimal set of parameters. However, this method may not be universally applicable to all materials, and it is recommended to limit the total number of iterations to a maximum of 100 to balance computational efficiency.

To guide the reader, the type of charge mixing algorithm used in Tables 1 and 2 below is determined by the IMIX parameter. Specifically, when IMIX is set to 1, the Kerker mixing algorithm is used; when IMIX is set to 2, the Tchebycheff mixing algorithm is applied. For IMIX equal to 4, the choice of the mixing algorithm depends on the weight factor WC: if WC is equal to 0, Broyden's method is used, while if WC is greater than 0, Pulay's mixing algorithm is chosen.

### 3. Results and Discussions

To evaluate the proposed approach, a diverse set of semiconducting, metallic, and insulating systems were selected as proof-of-concept examples. The crystal structures of the materials are provided in the supplementary materials. Reference calculations were performed using the default charge mixing parameters in the VASP code. These results serve as a basis for assessing the accuracy of the proposed approach in optimizing charge mixing parameters.

The first numerical experiment was conducted using the Superposition of Atomic Densities (SAD) method as an initial guess for the charge density. The Self-Consistent Iteration (SCI) count and CPU time for the systems studied are presented in Table 1, comparing the performance of default SAD (def. SAD) and optimized SAD (opt. SAD) parameters. Both the default and optimized parameters yielded the same energy with the desired SCF accuracy. However, the optimized parameters resulted in reductions in SCI by 13% (for LiBi) to 48% (for graphene), corresponding to decreases in single-point simulation time ranging from 7% (for TiMn) to 29% (for the Ni<sub>15</sub> cluster).

**Table 1:** SCI counts and CPU time for the systems studied using default (def. SAD) and optimized (opt. SAD) parameters, with the Superposition of Atomic Densities (SAD) method as the starting charge density guess.

		IMIX	AMIN	AMIX	BMIX	NELM DL	WC	MIX- PRE	MAX- MIX	SCI count	time
Graphene	def. SAD	4	0.10	0.4	1.00	-5	1000	1	-45	19	11.505
	opt. SAD	4	0.59	0.54	0.01	-2	661	2	-67	10	8.947
WS <sub>2</sub>	def. SAD	--	--	--	--	--	--	--	--	26	61.526
	opt. SAD	4	0.31	0.46	1.56	-2	1000	1	-45	17	44.786
Yb <sub>4</sub> Sb <sub>3</sub>	def. SAD	--	--	--	--	--	--	--	--	32	606.501
	opt. SAD	4	0.79	0.65	1.99	-3	1000	1	-45	22	441.301
Ni <sub>15</sub> cluster	def. SAD	--	--	--	--	--	--	--	--	90	394.248
	opt. SAD	4	0.18	0.24	0.50	-3	1000	1	-45	58	281.200
Cr <sub>15</sub> cluster	def. SAD	--	--	--	--	--	--	--	--	46	106.800
	opt. SAD	4	0.55	0.63	0.38	-4	1000	1	-45	34	86.832
SiC	def. SAD	--	--	--	--	--	--	--	--	17	60.669
	opt. SAD	4	0.60	0.54	1.89	-3	716	1	-194	13	48.728
DyPd	def. SAD	--	--	--	--	--	--	--	--	16	19.889
	opt. SAD	4	0.74	0.61	1.27	-2	806	0	175	13	17.917
AlNi	def. SAD	--	--	--	--	--	--	--	--	17	11.545
	opt. SAD	4	0.70	0.95	1.65	-3	690	1	197	13	10.683
Na	def. SAD	--	--	--	--	--	--	--	--	11	14.084
	opt.	1	--	0.93	0.24	-3	--	--	--	7	11.962

	SAD											
TiGa	def.	--	--	--	--	--	--	--	--	18	16.071	
	SAD											
	opt.	4	0.31	0.90	1.41	-2	578	1	192	13	13.660	
GaN	SAD											
	def.	--	--	--	--	--	--	--	--	16	22.742	
	opt.	4	0.24	0.95	1.72	-3	170	1	9	13	20.170	
PbLi	SAD											
	def.	--	--	--	--	--	--	--	--	13	11.488	
	opt.	2	1.84	0.67	0.62	-2	--	--	--	10	10.297	
LiBi	SAD											
	def.	--	--	--	--	--	--	--	--	15	42.250	
	opt.	4	1.07	0.08	0.07	-4	726	1	-152	13	38.093	
CaTl	SAD											
	def.	--	--	--	--	--	--	--	--	15	19.344	
	opt.	1	--	0.76	0.71	-3	--	--	--	12	16.695	
AlP	SAD											
	def.	--	--	--	--	--	--	--	--	16	59.672	
	opt.	4	0.57	0.62	0.67	-4	548	0	158	13	50.134	
Ca	SAD											
	def.	--	--	--	--	--	--	--	--	13	43.760	
	opt.	1	--	0.79	0.69	-3	--	--	--	10	34.835	
AlCuO <sub>2</sub>	SAD											
	def.	--	--	--	--	--	--	--	--	18	68.016	
	opt.	4	0.25	0.72	0.88	-3	54	0	-134	15	61.137	
Acetic acid molecule	SAD											
	def.	--	--	--	--	--	--	--	--	26	32.355	
	opt.	4	0.52	0.88	1.36	-5	144	2	-127	20	26.381	

Cu <sub>2</sub> O	def.	--	--	--	--	--	--	--	--	18	34.942
	SAD										
	opt.	4	0.02	0.74	1.58	-3	381	2	-125	15	31.352
	SAD										
NbMn	def.	--	--	--	--	--	--	--	--	17	15.431
	SAD										
	opt.	4	0.20	0.50	1.19	-3	512	2	58	14	14.088
	SAD										
KMgH <sub>3</sub>	def.	--	--	--	--	--	--	--	--	14	18.190
	SAD										
	opt.	4	0.71	0.86	0.56	-2	320	2	199	11	15.938
	SAD										
TiMn	def.	--	--	--	--	--	--	--	--	18	12.387
	SAD										
	opt.	4	0.27	0.55	1.81	-2	483	1	163	14	11.543
	SAD										

The second numerical experiment was conducted by using the charge density obtained from a previous calculation. In this case, a lower energy cut-off plane-wave calculation of 200 eV was performed, and the resulting charge density was used as the initial guess for a higher-energy cut-off calculation of 520 eV (CHG). The SCI count and CPU time for the systems studied are presented in Table 2, comparing default parameters using the 200 eV charge density as the initial guess (def. CHG), previously optimized parameters based on the SAD method (opt. SAD), and optimized parameters with the 200 eV charge density as the initial guess (opt. CHG).

From the results, we observe that in 17 cases from Table 2, the optimized parameters (opt. SAD), when starting from a coarser charge density, either achieve better convergence or yield the same SCI count compared to the default parameters. The corresponding time reduction ranges from 0% (for SiC) to 27% (for graphene). However, for two cases—AlNi and GaN—the default parameters converge more effectively than the optimized SAD ones. The best performance is observed when the optimization is performed with respect to the initial charge density guess, with SCI reduction ranging from 7% (for DyPd) to 43% (for graphene), and the corresponding reduction in CPU time ranges from 6% to 28%.

**Table 2:** SCI count and CPU time for the systems studied using default (def. CHG), optimized parameters based on SAD (opt. SAD), and optimized parameters using 200 eV charge density as the initial guess (opt. CHG).

		IMIX	AMIN	AMIX	BMIX	NELM DL	WC	MIX- PRE	MAX- MIX	SCI count	time
Gra- phene	def. CHG	4	0.10	0.4	1.00	-1	1000	1	-45	16	12.830
	opt. SAD	--	--	--	--	--	--	--	--	10	9.261
	opt. CHG	4	0.55	0.51	0.08	-1	890	2	93	9	9.227
WS <sub>2</sub>	def. CHG	--	--	--	--	--	--	--	--	22	69.544
	opt. SAD	--	--	--	--	--	--	--	--	15	49.934
	opt. CHG	4	0.84	1.13	1.48	-1	124	1	36	15	51.546
SiC	def. CHG	--	--	--	--	--	--	--	--	14	49.458
	opt. SAD	--	--	--	--	--	--	--	--	14	49.221
	opt. CHG	4	0.63	1.04	1.20	-1	298	2	-186	11	41.400
DyPd	def. CHG	--	--	--	--	--	--	--	--	13	17.366
	opt. SAD	--	--	--	--	--	--	--	--	13	17.969
	opt. CHG	4	0.82	1.23	1.88	-1	29	1	-92	12	16.321
AlNi	def. CHG	--	--	--	--	--	--	--	--	12	9.989
	opt. SAD	--	--	--	--	--	--	--	--	14	10.549
	opt. CHG	4	0.31	0.53	0.86	-1	667	1	27	10	9.509
Na	def. CHG	--	--	--	--	--	--	--	--	9	11.524
	opt. SAD	--	--	--	--	--	--	--	--	8	12.190
	opt. CHG	4	0.43	1.54	0.37	-1	636	2	180	6	10.323
TiGa	def. CHG	--	--	--	--	--	--	--	--	14	14.010
	opt. SAD	--	--	--	--	--	--	--	--	14	13.824
	opt. CHG	4	0.74	0.56	1.40	-1	618	0	153	13	12.850

GaN	def. CHG	--	--	--	--	--	--	--	--	12	18.411
	opt. SAD	--	--	--	--	--	--	--	--	13	20.635
	opt. CHG	4	0.28	1.06	0.99	-1	761	1	106	11	17.841
PbLi	def. CHG	--	--	--	--	--	--	--	--	10	10.610
	opt. SAD	--	--	--	--	--	--	--	--	10	10.073
	opt. CHG	1	1.21	0.84	0.57	-1	--	--	--	6	8.592
LiBi	def. CHG	--	--	--	--	--	--	--	--	13	34.898
	opt. SAD	--	--	--	--	--	--	--	--	13	38.400
	opt. CHG	4	0.40	0.69	1.48	-1	947	0	-87	10	29.471
CaTl	def. CHG	--	--	--	--	--	--	--	--	12	15.872
	opt. SAD	--	--	--	--	--	--	--	--	10	15.519
	opt. CHG	4	0.02	0.59	1.81	-1	315	0	197	10	14.755
AlP	def. CHG	--	--	--	--	--	--	--	--	13	46.698
	opt. SAD	--	--	--	--	--	--	--	--	13	49.682
	opt. CHG	4	0.64	0.68	0.02	-1	459	1	-87	9	34.612
Ca	def. CHG	--	--	--	--	--	--	--	--	11	35.932
	opt. SAD	--	--	--	--	--	--	--	--	10	34.795
	opt. CHG	1	1.34	0.94	0.93	-1	--	--	--	6	23.223
Acetic acid molecule	def. CHG	--	--	--	--	--	--	--	--	22	29.788
	opt. SAD	--	--	--	--	--	--	--	--	20	25.523
	opt. CHG	4	0.56	0.44	0.43	-1	105	0	104	15	22.433
Cu <sub>2</sub> O	def. CHG	--	--	--	--	--	--	--	--	16	31.763
	opt. SAD	--	--	--	--	--	--	--	--	15	30.125
	opt. CHG	4	0.61	1.71	1.49	-1	480	0	-66	14	29.203
NbMn	def. CHG	--	--	--	--	--	--	--	--	13	14.663
	opt. SAD	--	--	--	--	--	--	--	--	13	13.720
	opt. CHG	4	0.13	1.15	1.86	-1	384	1	-107	12	13.307
KMgH <sub>3</sub>	def. CHG	--	--	--	--	--	--	--	12	16.840	

	opt. SAD	--	--	--	--	--	--	--	--	11	15.820
	opt. CHG	1	0.33	0.88	0.78	-1	--	--	--	9	14.074
	def. CHG	--	--	--	--	--	--	--	--	14	11.134
TiMn	opt. SAD	--	--	--	--	--	--	--	--	14	10.997
	opt. CHG	4	0.08	0.42	2.00	-1	395	0	-18	13	10.254

The third numerical experiment involved testing the previously optimized parameters on perturbed systems by performing atomic relaxation. For the atomic relaxation tests, a 5% volume expansion was applied to the relaxed structures, and five relaxation iterations were performed for each material. The relaxation time for a subset of the systems previously studied is presented in Table 3, comparing the performance of the default SAD (def. SAD), default CHG (def. CHG), optimized SAD (opt. SAD), and optimized CHG (opt. CHG) parameters.

By comparing the time differences between relaxation with the optimized SAD (opt. SAD) and default CHG (def. CHG), we observe noticeable time reductions in many cases, with reductions reaching up to 26% (for graphene and WS<sub>2</sub>). However, there are five cases where the relaxation time increased. When using the optimized CHG parameters (opt. CHG), time reductions were observed for all cases, with the largest reduction of 32% for WS<sub>2</sub>.

**Table 3:** Relaxation time and corresponding time reduction for some materials studied, using default (def. SAD, def. CHG) and optimized (opt. SAD, opt. CHG) mixing parameters.

		Time (s) relaxation	Time reduction (%)
Graphene	def. AS	45.823	--
	def. CHG	44.800	--
	opt. AS	37.015	17.38%
	opt. CHG	32.789	26.81%
WS <sub>2</sub>	def. AS	267.495	--
	def. CHG	268.632	--
	opt. AS	196.150	26.98%
	opt. CHG	183.203	31.80%

SiC	def. AS	257.658	--
	def. CHG	242.631	--
	opt. AS	213.947	11.82%
	opt. CHG	232.872	4.02%
AlNi	def. AS	42.387	--
	def. CHG	41.158	--
	opt. AS	42.062	-2.20%
	opt. CHG	40.327	2.02%
Na	def. AS	60.732	--
	def. CHG	58.105	--
	opt. AS	50.226	13.56%
	opt. CHG	52.954	8.86%
TiGa	def. AS	46.213	--
	def. CHG	44.474	--
	opt. AS	43.380	2.46%
	opt. CHG	43.113	3.06%
GaN	def. AS	39.613	--
	def. CHG	37.030	--
	opt. AS	37.665	-1.71%
	opt. CHG	36.439	1.60%
PbLi	def. AS	19.012	--
	def. CHG	17.506	--
	opt. AS	17.477	0.17%
	opt. CHG	15.279	12.72%
AlP	def. AS	90.312	--
	def. CHG	80.880	--
	opt. AS	77.441	4.25%
	opt. CHG	70.751	12.52%
Ca	def. AS	58.369	--

	def. CHG	48.868	--
	opt. AS	51.635	-5.66%
	opt. CHG	47.496	2.81%
	def. AS	32.754	--
KMgH <sub>3</sub>	def. CHG	31.375	--
	opt. AS	32.562	-3.78%
	opt. CHG	29.130	7.16%

In the next step, the optimized mixing parameters were applied to various systems across different supercell sizes. Table 4 presents the SCI count for different materials and supercell sizes, using both the default (def. SAD) and optimized (opt. SAD) charge density mixing parameters. For most systems, the optimized parameters resulted in a significant reduction in SCI count, demonstrating faster convergence and more efficient computational performance. For example, in graphene, the SCI count decreases from 21 to 13 for both the  $2 \times 2 \times 1$  and  $3 \times 3 \times 1$  supercells. Similarly, in WS<sub>2</sub>, the SCI count reduces from 27 to 18 for the  $2 \times 2 \times 1$  supercell and from 28 to 20 for the  $3 \times 3 \times 1$  supercell. This trend holds true across both metallic and non-metallic systems, suggesting that the optimized parameters are broadly applicable.

These parameters are particularly advantageous for simulations involving larger supercells, such as those for elastic properties and phonon dispersion. The reusability of the optimized parameters across different system sizes minimizes the need for repeated, computationally expensive optimizations, thereby improving the efficiency of high-throughput applications in extended or supercell systems.

**Table 4:** SCI count for materials studied with optimized mixing parameters applied to different cell sizes.

		SCI count
Graphene ( $2 \times 2 \times 1$ )	def. SAD	21
	opt. SAD	13
Graphene ( $3 \times 3 \times 1$ )	def. SAD	21

	opt. SAD	13
WS <sub>2</sub> (2×2×1)	def. SAD	27
	opt. SAD	18
WS <sub>2</sub> (3×3×1)	def. SAD	28
	opt. SAD	20
Yb <sub>4</sub> Sb <sub>3</sub> (2×1×1)	def. SAD	33
	opt. SAD	23
SiC (2×1×1)	def. SAD	18
	opt. SAD	15
DyPd (2×2×1)	def. SAD	18
	opt. SAD	14
DyPd (2×2×2)	def. SAD	21
	opt. SAD	16
TiGa (2×2×1)	def. SAD	17
	opt. SAD	14
CaTl (2×2×2)	def. SAD	18
	opt. SAD	15
AlNi (2×2×2)	def. SAD	17
	opt. SAD	16
Ca (2×1×1)	def. SAD	15
	opt. SAD	12
Na (2×2×1)	def. SAD	13
	opt. SAD	9
TiGa (2×2×2)	def. SAD	17
	opt. SAD	14
PbLi (2×2×2)	def. SAD	13
	opt. SAD	11
NbMn (2×1×1)	def. SAD	18
	opt. SAD	17

The optimized mixing parameters were further evaluated using two additional DFT functionals: the local density approximation (LDA) [34] and the meta-GGA functional R<sup>2</sup>SCAN [37–39]. As shown in Table 5, the optimized parameters consistently reduce the number of self-consistent iterations (SCI) required for both functionals compared to the default parameters, indicating a significant improvement in convergence efficiency.

These results confirm the transferability of the optimized mixing parameters across different DFT functional types, specifically for LDA and R<sup>2</sup>SCAN, and reinforce their adaptability across LDA, GGA, and R<sup>2</sup>SCAN functionals. This finding is particularly noteworthy for meta-GGA functionals like R<sup>2</sup>SCAN, as SCAN-type functionals are known to exhibit oscillatory potentials where the random phase approximation (RPA) is often less accurate. The demonstrated robustness of these parameters suggests their potential applicability to a broader range of functionals, optimizing convergence and computational efficiency even in challenging cases.

**Table 5:** SCI count for systems studied with LDA and R<sup>2</sup>SCAN functionals, showing the impact of optimized mixing parameters compared to the default settings.

		SCI count (LDA)	SCI count (R <sup>2</sup> SCAN)
Graphene	def. SAD	19	20
	opt. SAD	12	12
WS <sub>2</sub>	def. SAD	25	32
	opt. SAD	16	28
SiC	def. SAD	15	17
	opt. SAD	12	14
AlNi	def. SAD	14	20
	opt. SAD	13	17

Na	def. SAD	11	13
	opt. SAD	8	11
TiGa	def. SAD	15	16
	opt. SAD	11	13
GaN	def. SAD	15	16
	opt. SAD	12	13
PbLi	def. SAD	11	14
	opt. SAD	9	11
AlP	def. SAD	16	19
	opt. SAD	12	13
Ca	def. SAD	13	20
	opt. SAD	12	16
KMgH <sub>3</sub>	def. SAD	16	17
	opt. SAD	12	12

In the final stage of the study, we evaluated the impact of the optimized mixing parameters on the simulation of mechanical properties for a range of materials: graphene, WS<sub>2</sub>, Yb<sub>4</sub>Sb<sub>3</sub>, SiC, and DyPd. These materials were chosen due to the significant computational resources required to accurately determine their elastic constants. To ensure precise force calculations, it is essential to use a supercell structure of the primitive cell, as detailed in Table 6 for each material system [48,49]. We employed the energy-strain method [50], recognized for its high accuracy in calculating elastic constants, which involves applying multiple constraints to the system, each requiring a separate calculation [51,52].

The results confirm that the optimized mixing parameters are transferable to perturbed systems, maintaining their efficacy even when atomic positions are altered. Table 6 summarizes

the calculated elastic constants and corresponding simulation times (in seconds) for each material. The identical elastic constants obtained with both the default and optimized parameters verify the accuracy of the optimized approach. Subsequently, we analyzed the reduction in computational time achieved with the optimized parameters compared to the default settings. As shown in Table 6, the optimized parameters yielded time savings of 33%, 29%, 28%, 12%, and 12% for graphene, WS<sub>2</sub>, Yb<sub>4</sub>Sb<sub>3</sub>, SiC, and DyPd, respectively. These reductions represent significant energy savings, without any compromise in accuracy. For instance, the time saved for Yb<sub>4</sub>Sb<sub>3</sub> on a 48-core cluster translates to almost two hours of computational time conserved.

**Table 6:** Calculated elastic constants for Graphene, WS<sub>2</sub>, Yb<sub>4</sub>Sb<sub>3</sub> SiC and DyPd.

		$C_{11}$ (GPa)	$C_{12}$ (GPa)	$C_{44}$ (GPa)	CPU time (s)
Graphene (3×3×1)	def. SAD	352.311	64.013	--	891.606
	opt. SAD	--	--	--	598.333
WS <sub>2</sub> (2×2×1)	def. SAD	176.814	52.539	--	1390.344
	opt. SAD	--	--	--	985.509
Yb <sub>4</sub> Sb <sub>3</sub> (conventional cell)	def. SAD	96.137	19.833	25.531	19869.625
	opt. SAD	--	--	--	14316.619
SiC (conventional cell)	def. SAD	383.767	127.215	241.019	180.394
	opt. SAD	--	--	--	162.378
DyPd (2×2×2)	def. SAD	123.883	70.390	26.800	100.954
	opt. SAD	--	--	--	88.621

#### 4. Conclusion

In this study, we propose the use of Bayesian optimization to reduce the computational cost of density functional theory (DFT) simulations while preserving their accuracy. This approach has been demonstrated through benchmark calculations on a range of systems, including metallic, insulating, and semiconducting materials. The results show that the efficient optimization of charge-mixing parameters using the Bayesian algorithm significantly reduces simulation time by minimizing the number of self-consistent iterations required for convergence. Importantly, this reduction in computational effort does not compromise the accuracy of the results. Our method offers a straightforward and effective way to alleviate some of the computational burden of DFT calculations while maintaining their accuracy. This approach can be extended to a broad spectrum of materials and systems, making it a valuable tool for researchers in physics, chemistry, and nanoscience who rely on DFT simulations for their work.

**Authors Contributions:**

Mohammed Benaissa conceptualized the study, performed the DFT simulations, and drafted the manuscript. Tarik Ouahrani performed the calculations and contributed to the draft and manuscript preparation. Didier Sébilleau provided conceptual guidance throughout the study. Keisuke Hatada contributed computational resources and assisted with draft and manuscript preparation. Kazuki Yoshikawa assisted with performing the DFT simulations and contributed to the manuscript preparation. All the authors reviewed and approved the final version of the manuscript.

**Supporting Information**

Lattice structure and DFT simulation parameters used for each material.

**Acknowledgments**

- Mohammed Benaissa acknowledges partial funding from Horizon Europe MSCA Doctoral network grant n.101073486, EUSpecLab, funded by the European Union.
- This work is supported by "Le Centre National de la Recherche Scientifique (CNRS)"
- This work is supported by "La Direction Générale de la Recherche Scientifique et du Développement Technologique (DGRSDT) "

**Competing interests:**

The authors declare that they have no competing interests.

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**Data availability**

The data are provided with this paper. Additional data that support the findings of this study are available from the corresponding author upon request.

**Code availability**

The code that supports the findings of this study is available from the corresponding author upon request.

Journal Pre-proof

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**Declaration of interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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