Optimizing DFT Hybrid Functionals for 2D Materials Using Genetic Algorithms

María Camarasa-Gómez[1], Daniel Sánchez-Portal[1], Cristian Ramirez-Atencia[2]
[1]Centro de Física de Materiales CFM/MPC (CSIC-UPV/EHU), Paseo de Manuel Lardizabal 5, Donostia-San Sebastián 20018, Spain
[2]Universidad Politécnica de Madrid, C/ Alan Turing, s/n, Madrid 28031, Spain

maria.camarasa@ehu.eus

In recent years, there has been a widespread interest in the determination of electronic structure and optical absorption spectra of two-dimensional (2D) van der Waals (vdW) materials due to their vast potential of being crucial elements of next-generation optoelectronic and quantum technologies material components. However, predicting these properties from *ab initio* has proved to be an outstanding challenge. In recent years, the use of hybrid functionals within the time-dependent density functional theory framework has emerged as a serious contender to GW and the Bethe-Salpeter equation (BSE) calculations [1, 2, 3], which are widely regarded as the benchmark methods for accurate predictions in the field. However, the performance of these functionals is highly sensitive to the value of the fraction of exact exchange (α) and the range-separation parameter (γ) considered. Traditional methods for tuning these parameters are computationally expensive and lack flexibility when applied to different materials.

This work proposes a genetic algorithm (GA)-based framework [4] for optimizing α and γ in order to improve the accuracy and efficiency of hybrid functionals in modeling the electronic properties of 2D vdW materials. The GA exploits principles of evolutionary computation to explore the hyperparameter space efficiently, balancing accuracy with computational cost. Validation is performed for prototypical vdW materials. We compare with GW electronic structure calculations, demonstrating that the GA-tuned parameters yield accurate electronic structure results. Our findings reveal that our approach reduces the trial-and-error workload thus accelerating the characterization of more complex structures, paving the way for automated and adaptive tuning strategies in computational materials science.

References

[1] A. Ramasubramaniam, D. Wing, and L. Kronik, *Phys. Rev. Mater.* **3,** 8 (2019) 084007.
[2] M. Camarasa-Gómez, A. Ramasubramaniam, J. B. Neaton, and L. Kronik, *Phys. Rev. Mater.* **7**, 10 (2023) 104001.
[3] M. Camarasa-Gómez, S. E. Gant, G. Ohad, et al., *npj Comput. Mater.* **10**, (2024) 288
[4] J. Blank and K. Deb, *IEEE Access* **8**, (2020) 89497.

Figures

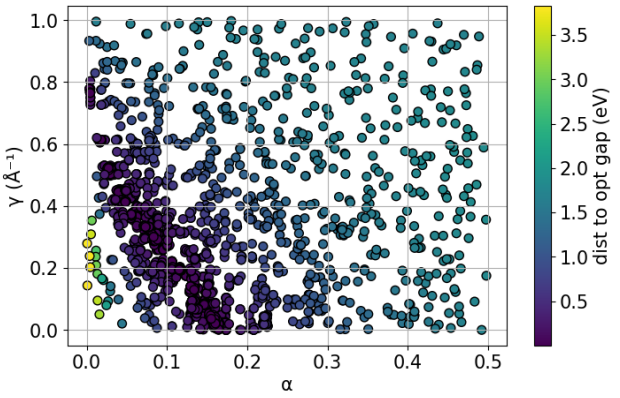

Figure 1. Sample of contour map of the hyperparameter space search generated by the GA-algorithm in the optimization of the exact exchange (α) and the range separation parameter (γ) for monolayer MoS2. Each point represents a potential solution generated by the GA-algorithm, with the color indicating the distance to the optimized target, the reference band gap.