From structure to property and back

Ivor Lončarić [1, †]

[1]Ruđer Bošković Institute, Bijenička cesta 54, 10000 Zagreb, Croatia
†corresponding author's email: ivor.loncaric@irb.hr

Machine-learning interatomic potentials (MLIPs) are often introduced simply as faster surrogates for first-principles calculations. However, they change the type of questions that can be addressed computationally: from following complex finite-temperature phenomena in an individual material, through screening large materials spaces for unusual properties, to ultimately designing new structures with desired characteristics.

We have constructed MLIPs to extend first-principles modelling to spatial and temporal scales required to describe complex material behaviour. A particularly illustrative example is provided by thermosalient molecular crystals, which undergo phase transitions accompanied by large mechanical motion. There, MLIP enabled Gibbs free-energy calculations that reproduce the experimentally observed phase transition and extreme negative thermal expansion, and revealed the elastic origin of the barrier associated with the transition [1]. Combining experiment and MLIP simulations subsequently revealed an extreme negative linear compressibility of approximately $-39\ \mathrm{TPa}^{-1}$ and its unusual pressure-dependent switching [2]. These examples illustrate how MLIPs can provide atomistic interpretation of experimentally observed functional responses that would be difficult to access through direct first-principles simulations.

The next step is to move from individual materials to materials spaces. We developed global MLIPs for molecular crystals by exploiting existing molecular and crystalline first-principles datasets [3] and we benchmarked universal MLIPs for elastic properties and used them for high-throughput screening of molecular crystals, identifying candidates with unusual mechanical responses, including negative linear compressibility, which were subsequently validated using density-functional theory [4]. Thus, sufficiently transferable atomistic models transform property calculation into a scalable search problem.

The natural final step is to reverse the direction: instead of asking only *what properties does this structure have?*, we would like to efficiently explore structures that have not yet been considered. Crystalite is a lightweight diffusion Transformer for crystal structure prediction and de novo crystal generation, combining chemically structured subatomic tokenization with geometry-aware attention [5]. It achieves strong generative performance while sampling substantially faster than geometry-heavy equivariant architectures. Combining such generative models with rapid MLIP-based relaxation and property evaluation provides a route towards a closed computational discovery loop: *structure* → *property* → *new structure*.

Acknowledgments

I acknowledge COST Action CA22154 - Data-driven Applications towards the Engineering of functional Materials: an Open Network (DAEMON) supported by COST (European Cooperation in Science and Technology), the European Regional Development Fund for the project 'Materials for clean energy, advanced sensors and quantum technologies' (Grants No. PK.1.1.10.0002), Croatian Science Foundation for the project IP-2026-5909, and support by the European Union's NextGenerationEU program.

References

[1] B. Mladineo and I. Lončarić, *Crystal Growth Design* **24**, 8167–8173 (2024).
[2] B. Mladineo, T. Klaser, M. Ende, J. Popović, I. Lončarić, and Ž. Skoko, *Crystal Growth Design* **25**, 8196–8202 (2025).
[3] I. Žugec, R. M. Geilhufe, and I. Lončarić, *Journal of Chemical Physics* **160**, 154106 (2024).
[4] A. Kholtobina and I. Lončarić, *Materials & Design* **254**, 114047 (2025).
[5] T. Hadži Veljković, J. Rosenthal, I. Lončarić, and J.-W. van de Meent, *arXiv:2604.02270* (2026).