

NUEVO!

Potenciales entrenados por Machine Learning (MLiPS)nature  
COMMUNICATIONS

ARTICLE

DOI: 10.1038/s41467-018-06169-2

OPEN

# Towards exact molecular dynamics simulations with machine-learned force fields

Stefan Chmiela<sup>1</sup>, Huziel E. Sauceda<sup>2</sup>, Klaus-Robert Müller<sup>1,3,4</sup> & Alexandre Tkatchenko<sup>5</sup>

Molecular dynamics (MD) simulations employing classical force fields constitute the cornerstone of contemporary atomistic modeling in chemistry, biology, and materials science. However, the predictive power of these simulations is only as good as the underlying interatomic potential. Classical potentials often fail to faithfully capture key quantum effects in molecules and materials. Here we enable the direct construction of flexible molecular force fields from high-level ab initio calculations by incorporating spatial and temporal physical symmetries into a gradient-domain machine learning (sGDML) model in an automatic data-driven way. The developed sGDML approach faithfully reproduces global force fields at quantum-chemical CCSD(T) level of accuracy and allows converged molecular dynamics simulations with fully quantized electrons and nuclei. We present MD simulations, for flexible molecules with up to a few dozen atoms and provide insights into the dynamical behavior of these molecules. Our approach provides the key missing ingredient for achieving spectroscopic accuracy in molecular simulations.

→ Buen modelo para vibraciones moleculares de alta energía.

→ Ab-Initio Molecular dynamics

- Propiedades electrónicas + dinámica de iones  
(DFT + MD = AiMD)

- Descripción cuántica de alta precisión
- Buen poder predictivo de propiedades materiales

⊗ Sistemas pequeños

⊗ Tiempos cortos

- Requerimiento computacional muy grande

- Dinámica Molecular: dinámica y termodinámica

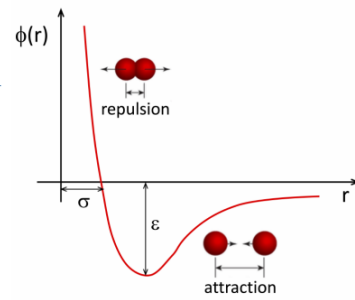
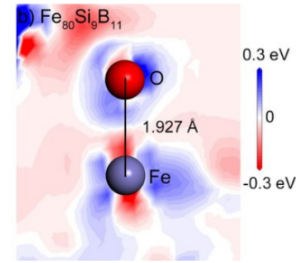
- Descripción con potenciales clásicos, precisión variable.

- Menor poder predictivo que Ab Initio - MD.

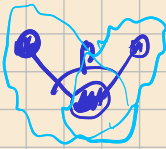
⊗ Sistemas de mayor tamaño

⊗ Tiempos largos (permite el cómputo de cantidades muy fluctuantes).

- Procesos fuera de equilibrio (flujo de líquidos, flujos térmicos)

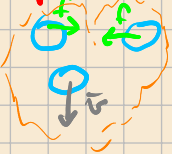


$$\phi(r) = 4\epsilon \left[ \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^6 \right]$$



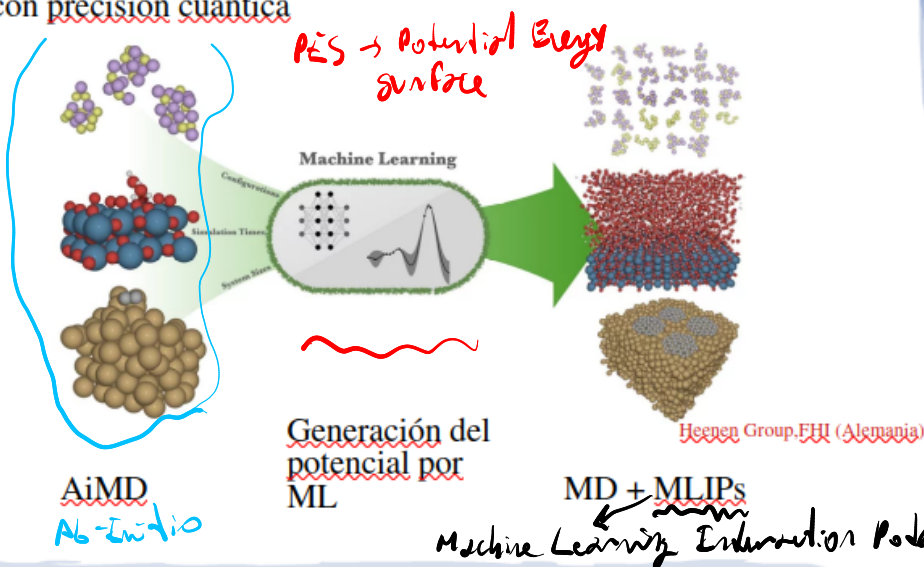
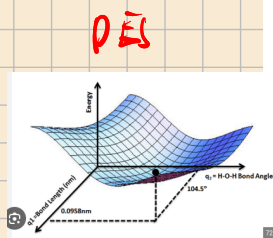
$$V = \frac{1}{2} \sum_{i,j} q_i q_j / r_{ij}$$

Many body problem



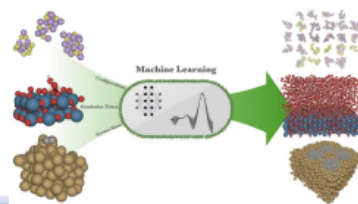
# Qué aporta Machine Learning?

- Permite unir los dos mundos
- Simulaciones de muchas partículas por tiempos largos con precisión cuántica



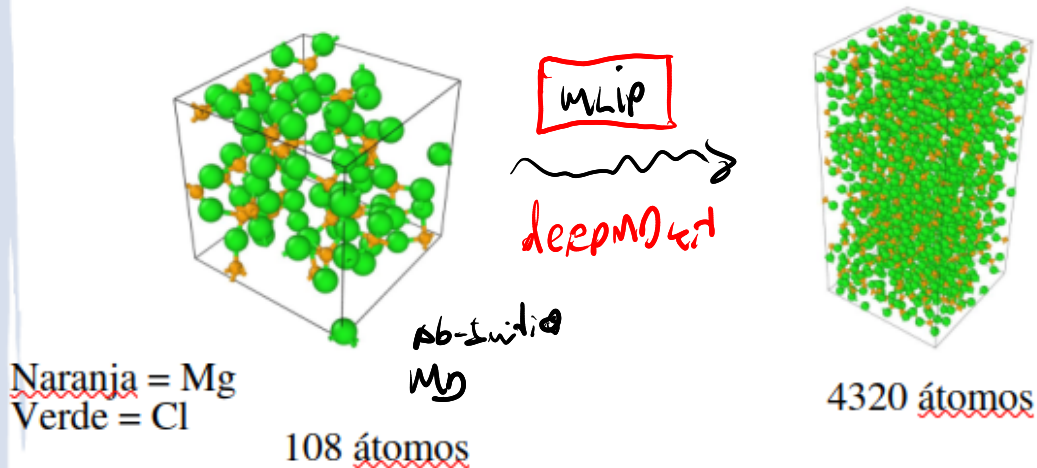
## Dinámica Molecular con potenciales MLIPs

- Precisión cuántica, comparable a AiMD
- Simulaciones de hasta millones de átomos
- Tiempos largos de simulación
- Gran poder predictivo para el cálculo de propiedades termodinámicas, dinámicas y estructurales
- Simulaciones fuera de equilibrio (flujos en reactores, intercambiadores de calor)
- Procesos interfaciales
- Propiedades físico-químicas en condiciones experimentales difíciles, costosas o imposibles
- Enriquecimiento de materiales nucleares (Li, U)



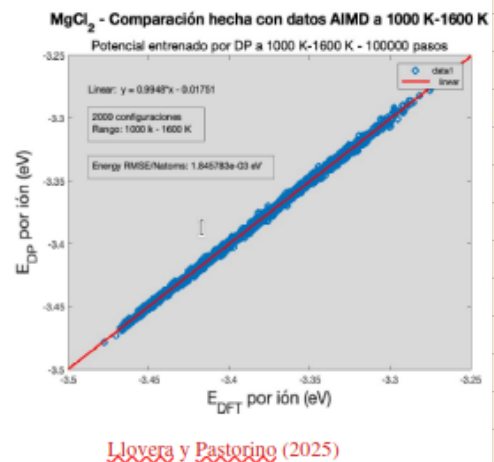
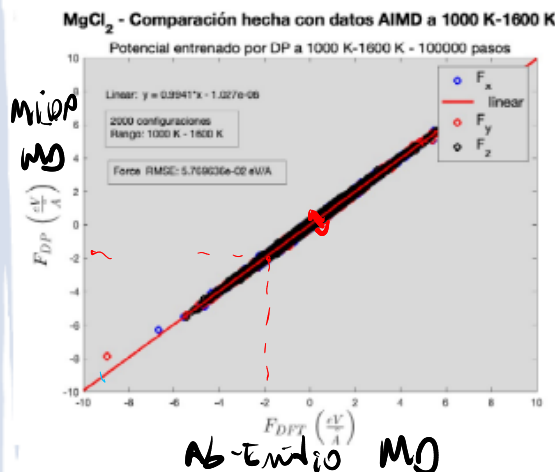
# Ejemplo de entrenamiento de MLIP

- Sales Fundidas a altas temperaturas (1000-1600K)
- Un ejemplo:  $\text{MgCl}_2$
- **Aplicación:** Reactores de Sal Fundida
- Conocimiento incompleto de propiedades físico-químicas
- Experimentos muy complejos: alta temperatura, control de impurezas, mitigación de la corrosión



## En CNEA: Sal de $\text{MgCl}_2$

- Estudio de sales fundidas cloradas y sus mezclas eutécticas ( $\text{MgCl}_2$ ,  $\text{NaCl}$ ,  $\text{KCl}$ )
- Simulaciones AiMD, red neuronal entrenada con 56000 configuraciones, se verifica con 7000 configuraciones (no usadas para entrenar)



# Densidad y Calor Específico

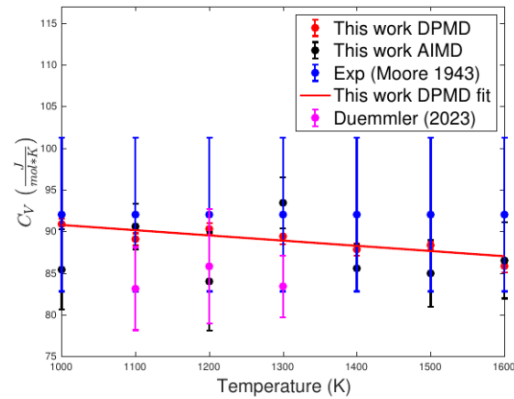
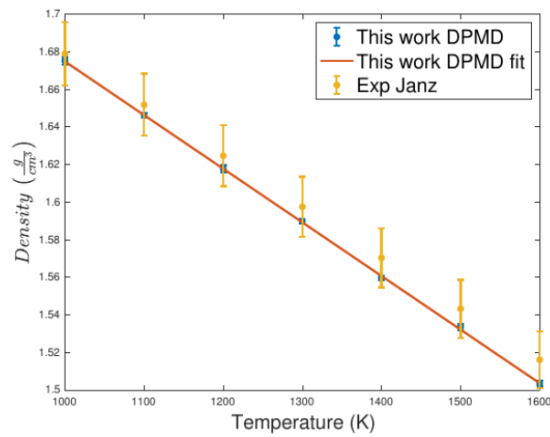


Figure 12. Heat capacity of molten  $\text{MgCl}_2$  in the 1000 K – 1600 K range, calculated by DPMD.

Llovera, Barral, Vildosola y Pastorino (En prep. 2025)

Ejemplo de muestra muy grande

## Conductividad térmica Au y Ag

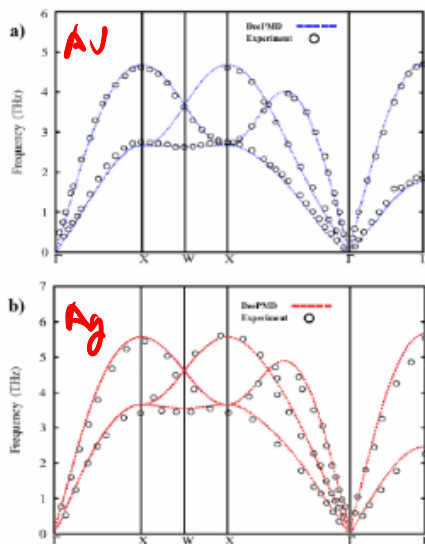


Figure 5: Computed and Experimentally-sourced phonon dispersion of (a) Au and (b) Ag crystal structures.

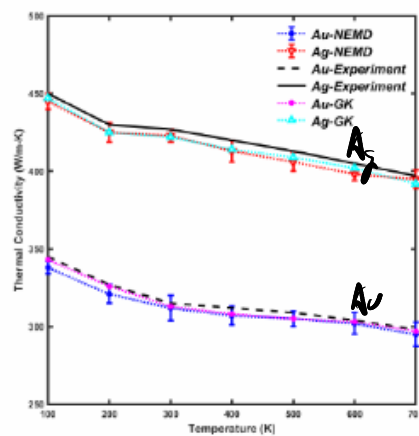


Figure 6: Computed thermal conductivity for Au and Ag systems at varying temperatures between 100-700K. Both NEMD and the GK methods produce accurate determinations of these values.

Piroozan & Kumar SC-W 2023

Muestra: 2M átomos

$$\omega = \sqrt{\frac{k}{m}}$$

$$J = -K \frac{\partial T}{\partial x}$$

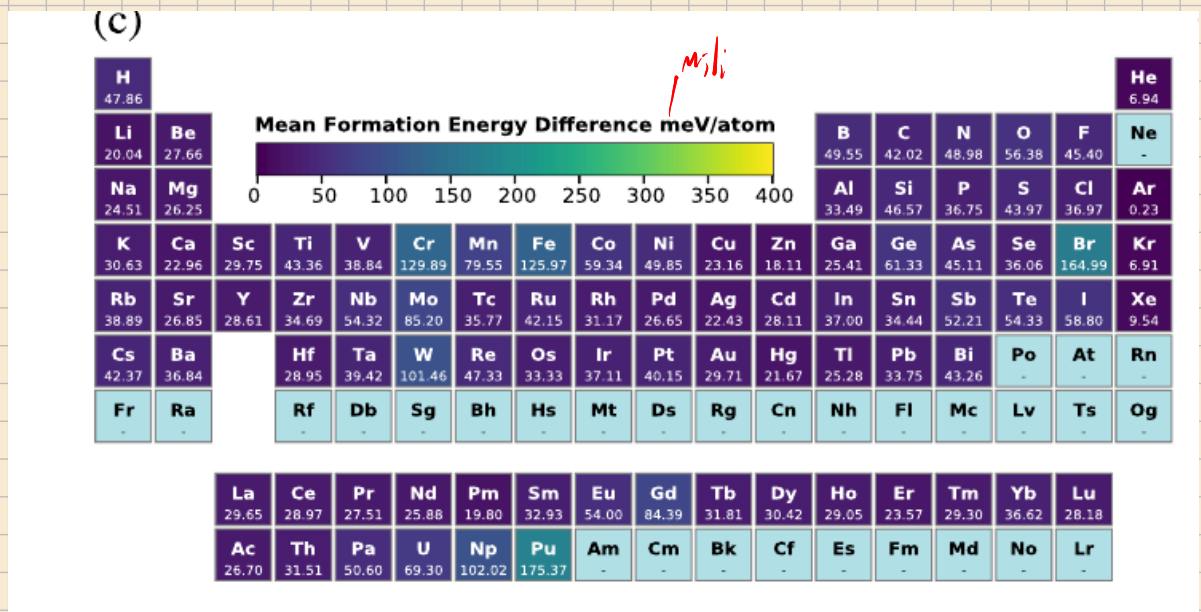
Conductividad térmica

# Foundation Models (uMLIPS)

- Potenciales MLIPs universales. (?)
- Entrenados con muchísimas trayectorias de AIMD.
- **Ejemplo: MACE.** Entrenado con Materials Project
  - MPTraj: 1.58M trayectorias.
  - 10+ años de cálculos DFT.
  - 154k identificadores de distintos materiales.
  - Mayoría de cristales bulk



## Foundation Models: comparison de energies de formation



Yu et. Al, MGE Advances 2:e58 (2024)

$$E_{\text{form}}[A_aB_b] = E[A_aB_b] - x_a E[A] - x_b E[B], \quad (3)$$

where  $E[A_aB_b]$  is the total energy per atom of the phase of interest with  $A$ ,  $B$  the constituent elements,  $x_a = a/(a + b)$  and  $x_b = b/(a + b)$  are the fractions of  $A$  and  $B$ , respectively, and  $E[A]$  and  $E[B]$  are the lowest possible energies per atom of  $A$  and  $B$ , respectively. The ability of a uMLIP in predicting formation energies is evaluated as a difference from the MP values, *that is* as follows:

$$\Delta E_{\text{form}} = E_{\text{form}}^{\text{MP}} - E_{\text{form}}^{\text{uMLIP}}, \quad (4)$$

<https://onlinelibrary.wiley.com/doi/epdf/10.1002/mgea.58>

MACE-MP-0: *submitted* of <https://arxiv.org/pdf/2401.00096>

# MACE: Reentrenamiento

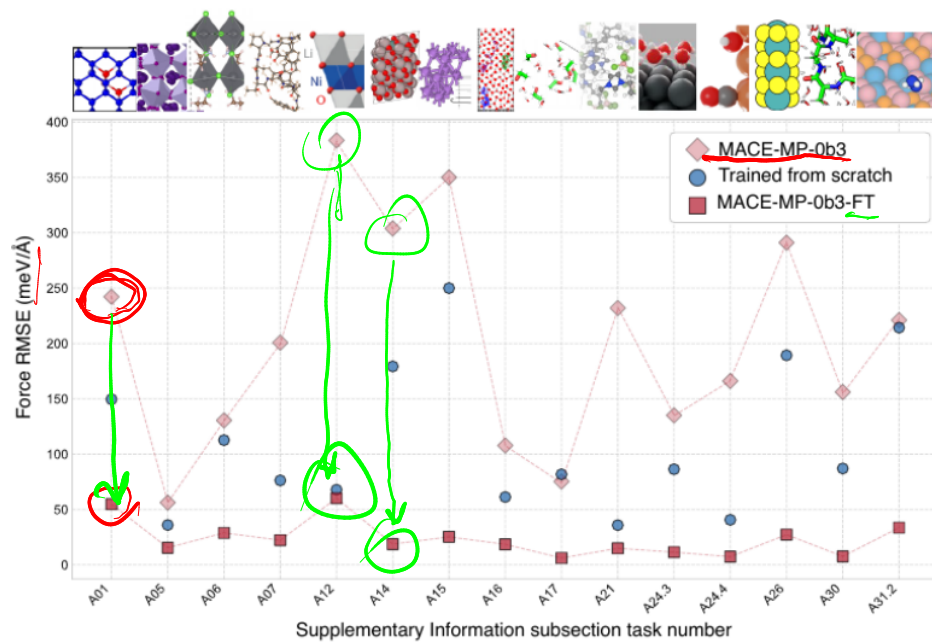


Figure 4: **Fine-tuning.** A comparison of force RMS error on selected applications in the SI for which fine-tuning was performed. The MACE-MP-0b3 model is shown with pink diamonds and the fine-tuned model (MACE-MP-0b3-FT) for each application with red squares. For comparison, in each case we also show the results corresponding to a model trained “from scratch” only to the small amount of fine-tuning data (blue circles).

Batatia et al. <https://arxiv.org/pdf/2401.00096> (2025)