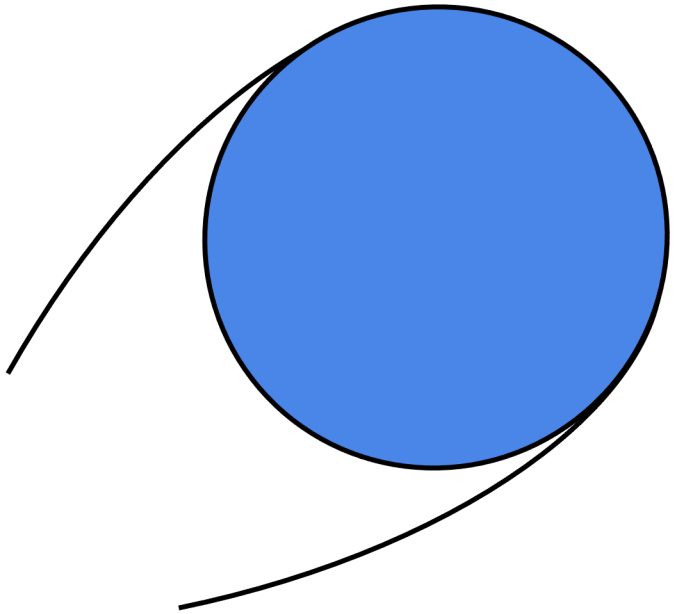


Machine learning based analysis of collective diffusion in inorganic solid-state electrolytes

Cibrán López Álvarez[1,2,3] Riccardo Rurali[3]

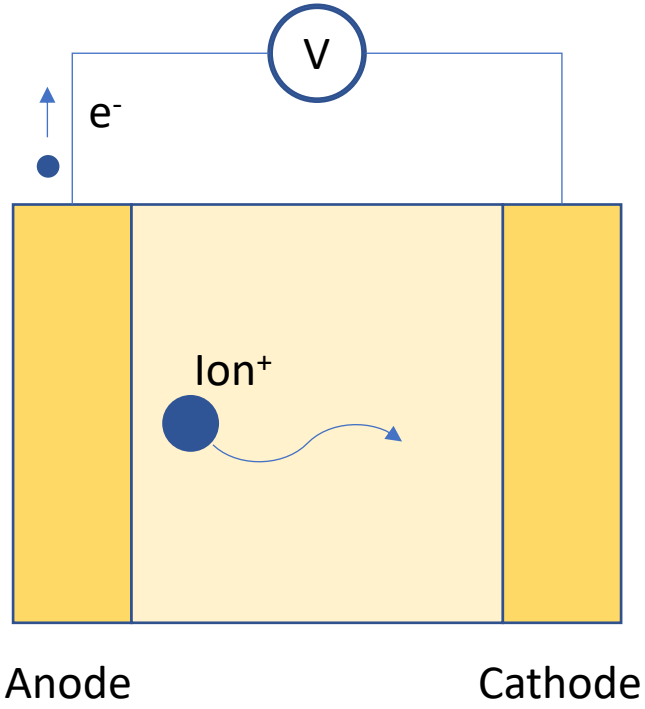
Claudio Cazorla Silva[1,2]

[1]Departament de Física, UPC

[2]Barcelona Research Center in Multiscale Science and Engineering, UPC

[3]Institut de Ciència de Materials de Barcelon, ICMAB-CSIC, UAB

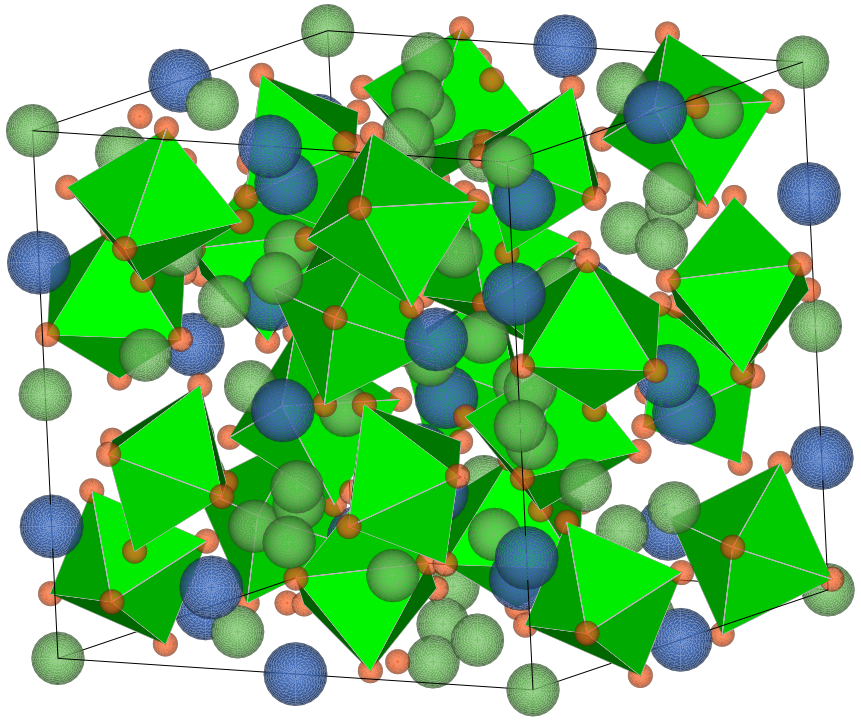
What are solid-state batteries?



All-solid-state batteries consist on a new generation of energy storage devices which present high stability and safety.

In a broad sense, the diffusion of cations within the solid electrolyte from anode towards cathode generates an external voltage.

LLZO as an example

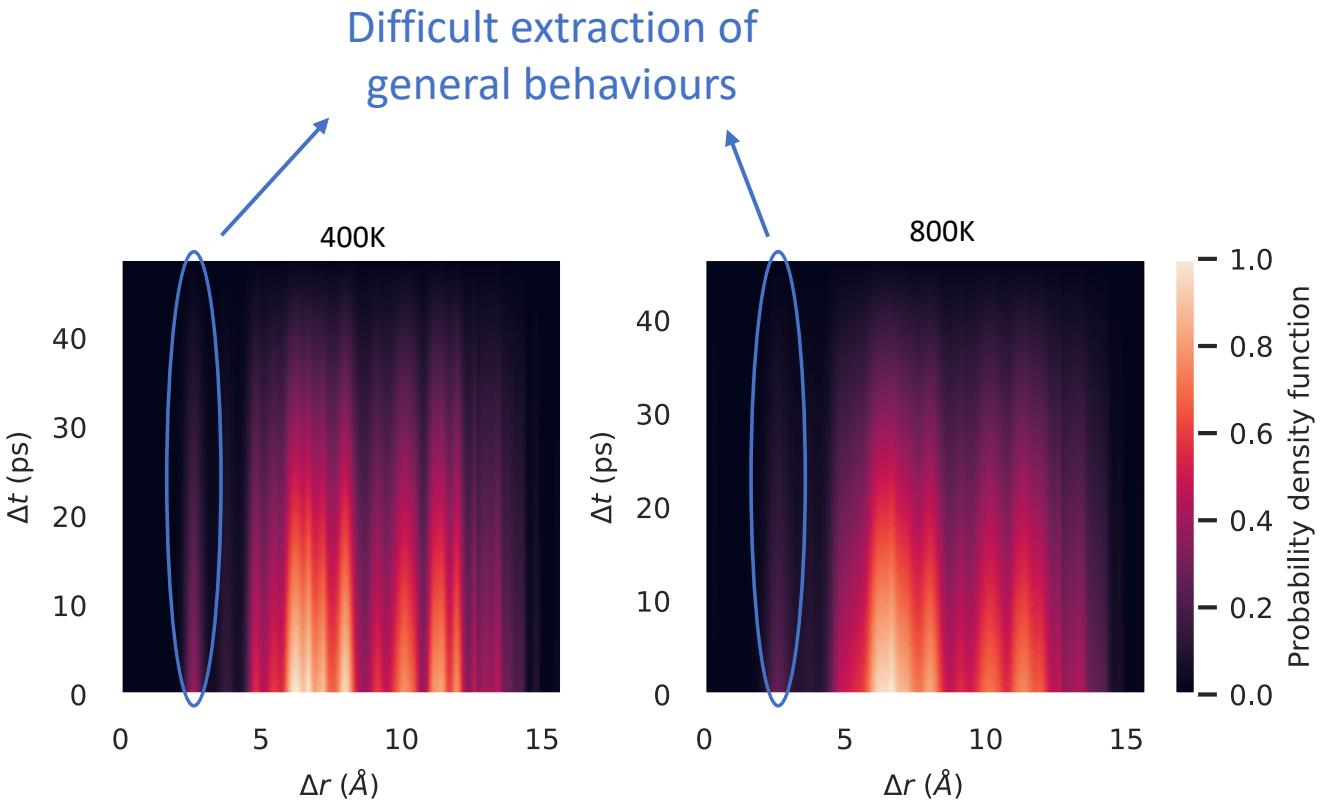


Most common fast-ion conductors typically rely on Lithium (or more recently Sodium as well) diffusion.

The Li-garnet electrolyte $Li_7La_3Zr_2O_{12}$ consists on a canonical example of conductor, exhibiting low electrical conductivity and wide electrochemical operation window, with a conductivity of up to 1 S/cm at room temperature.

However, outperforming current benchmarks requires a deeper understanding of processes governing ion diffusion.

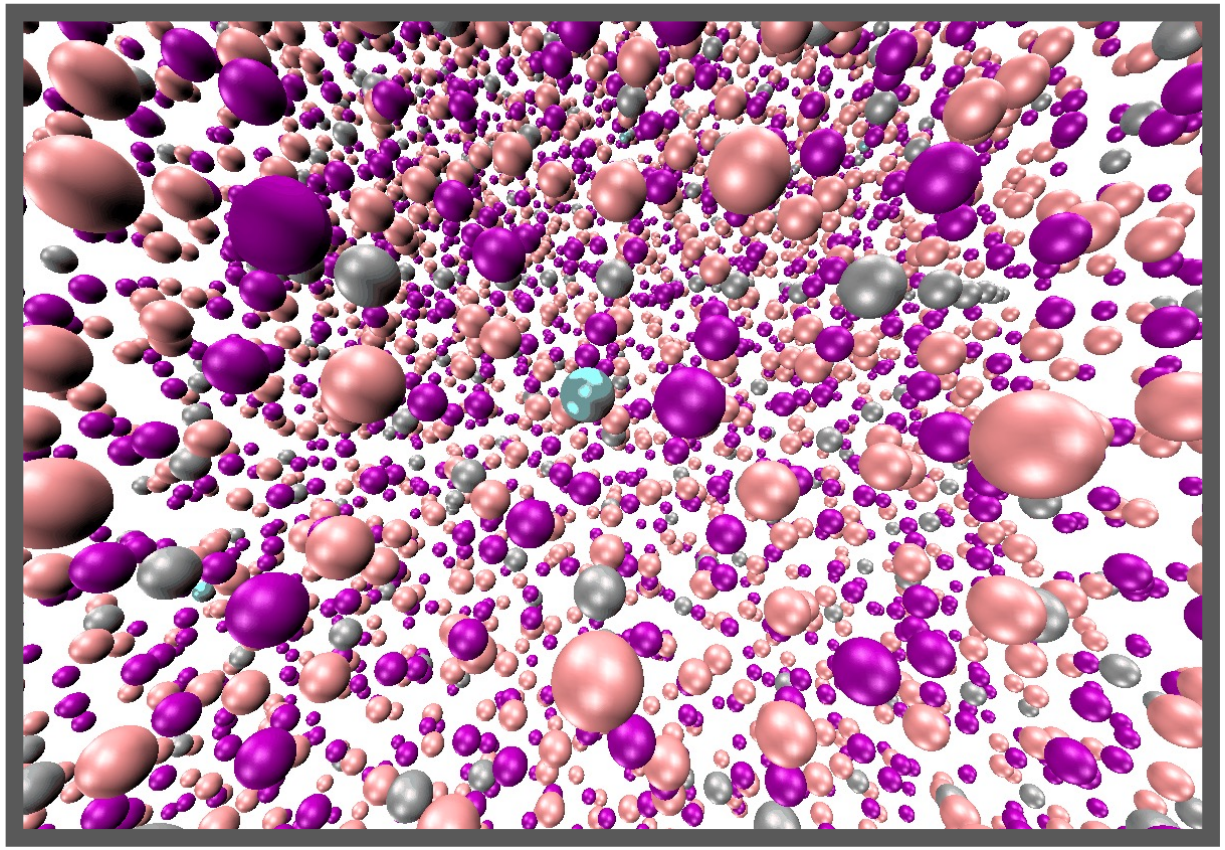
Previous approaches



Previous approaches rely in either arbitrarily defined geometrical parameters for identifying hopping events, or complex spatio-temporal correlation distributions such as the van Hove's correlation function.

Unsupervised algorithms would enable systematic and more rigorous studies of diffusion processes, skipping the definition of any material-dependent parameter.

How does a simulation look like?

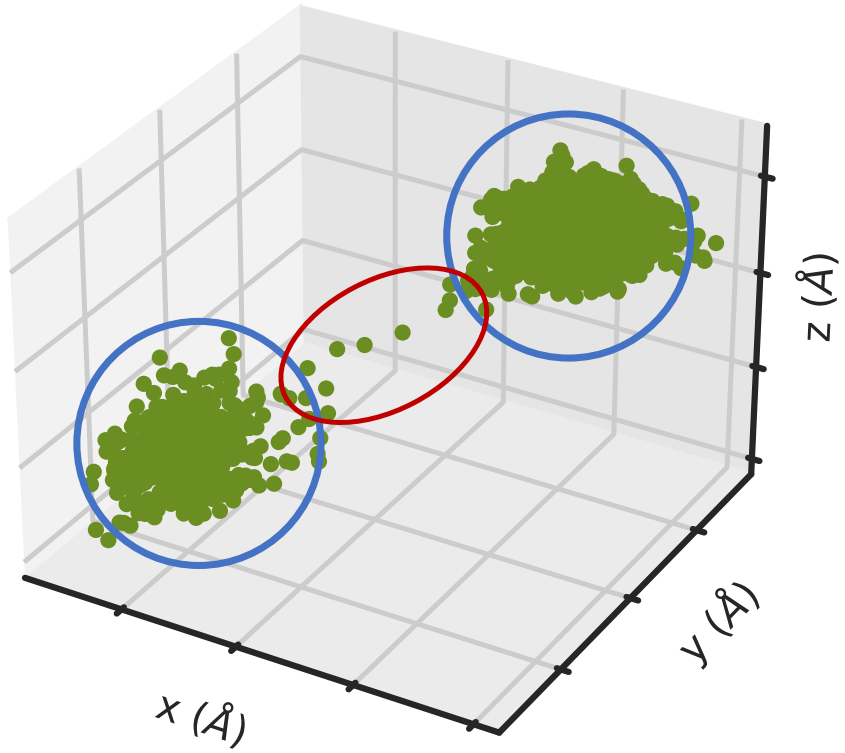


Diffusive Lithium particle

This AIMD simulation of LLZO shows the diffusion of a particle (light blue).

Apart from the rather few time steps in which this process takes place, most ions just vibrate around a steady point.

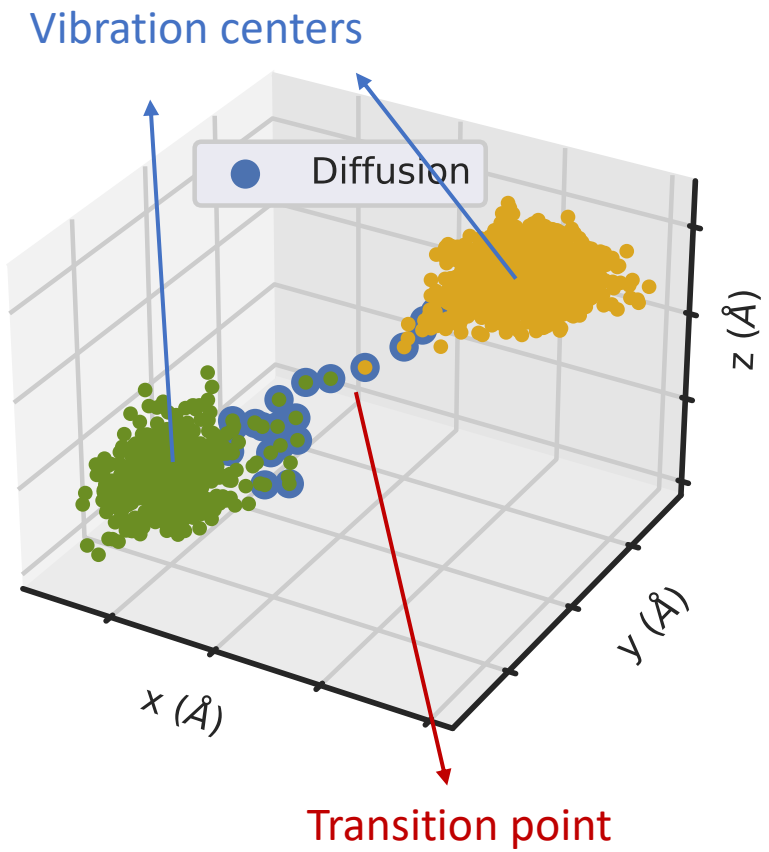
Trajectory of a single particle



Let’s isolate the trajectory of a single particle (concretely, the diffusive one from the previous simulation).

From this, the vibrating and diffusive sections can be easily spot.

K-means approach

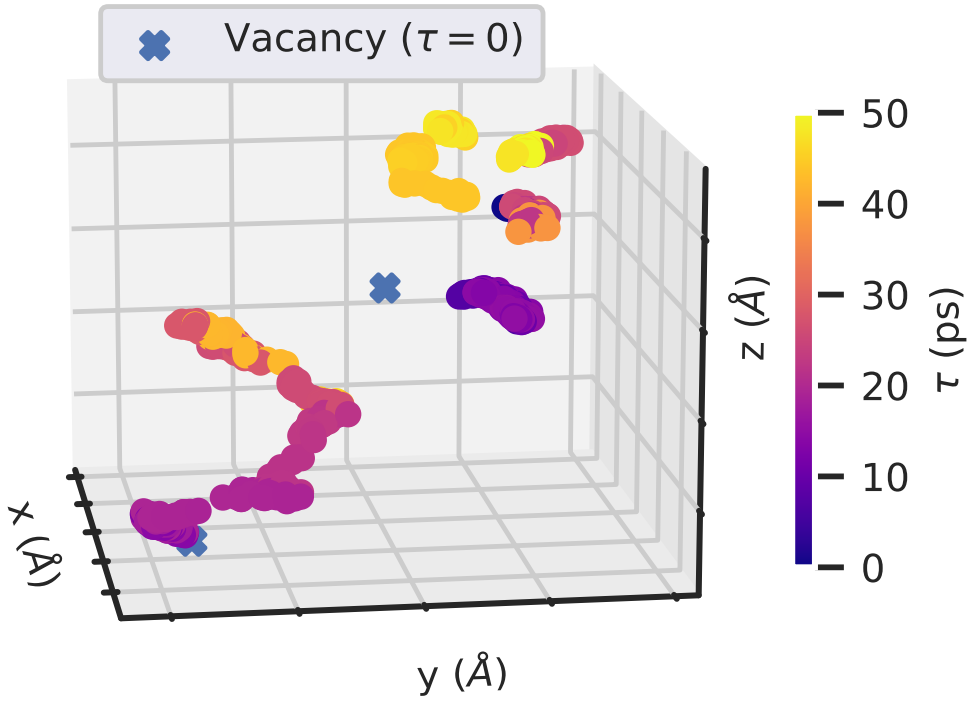


Clustering algorithms, as part of unsupervised machine learning models, are devoted to extracting unlabeled trends from data.

Here we use it to identify vibrational clusters, thus distributed clustering might be preferred (K-means, for example).

Diffusive sections are defined as the connection between vibrational clusters. Density-based algorithms (such as spectral clustering) were also studied, misperforming.

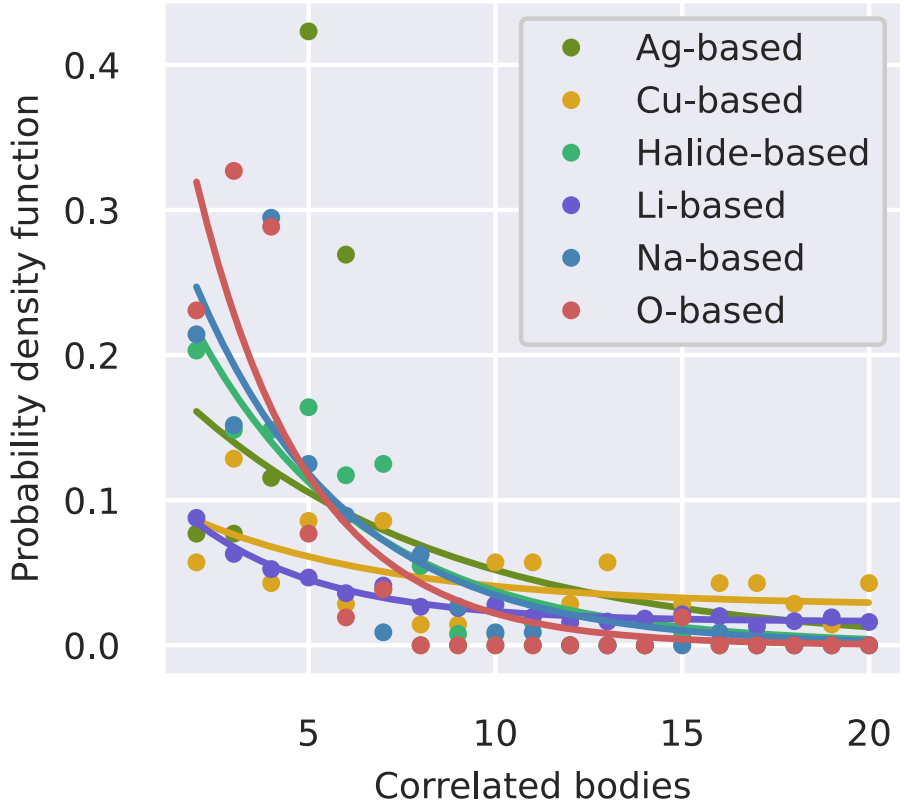
K-means approach



Repeating this identification for each particle in a simulation, we can extract all the diffusive paths and represent them in tems of simulation time.

It is straightforward to visually check some trends in diffusion events (chain-like events), as well as a close relation between vacancy position and evolution of the diffusion processes.

Analysis of correlations



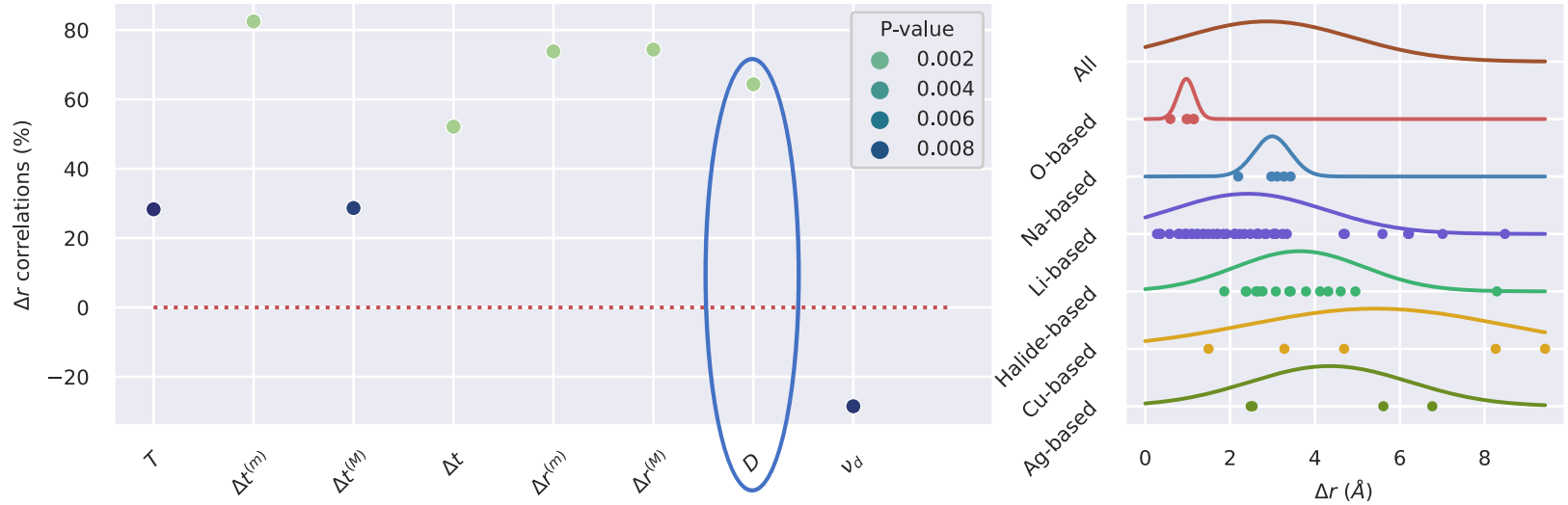
Each family presents the same trend, of exponential decay of the correlation with the number of bodies involved in the correlation.

This means that, although chain-like processes are the most important in these elements, considering the interaction of more distant particles might be of relevance as well when designing new candidates.

Analysis of descriptors

This approach allows studying any possible atomistic descriptor, such as spatial and temporal lengths of the diffusive events, and their correlation with parameters (diffusion coefficient, for example).

Such a reasearch line could lead to understading how atomistic descriptors balance each other (thus,to physical-informed search of new materials).



Overlook

Unsupervised machine learning models are an efficient tool for extracting information in computational chemistry, with growing interest considering the already existing datasets.

Our specifc approach allowed, for the first time:

- Quantifying significative many-body correlations, pivotal as well for correctly computing the diffusion coefficient.
- Accessing atomistic descriptors defining diffusion events, that can be used for an efficient search of novel candidates.

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BARCELONATECH

Reach out at:

cibran.lopez@upc.edu

GitHub repository:

github.com/IonRepo/IonDiff