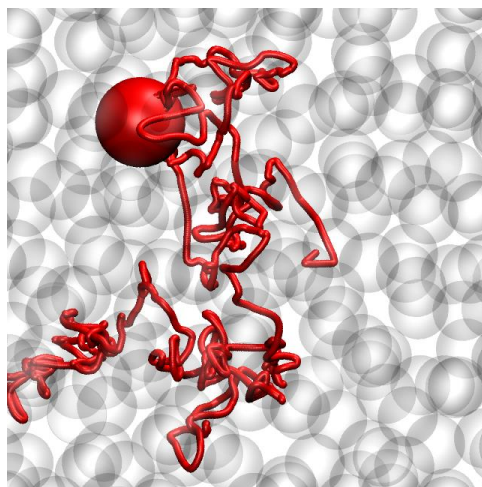




PISACMS 2017

August 28 – Sep. 01
Paris, Campus Jussieu





Carried by Sorbonne Universities, LabEx MATISSE (MATERIALS, Interfaces, Surfaces, Environment) is a multidisciplinary project on the borders of the chemistry, physics and Earth sciences and mobilizes a critical mass of research laboratories around the study of materials, so natural as synthetic, in the service of big socioeconomic stakes. It federates 18 prestigious actors around interdisciplinary projects.

MATISSE gathers exceptional skills regarding elaboration of materials by chemical or physical ways, characterization of the properties since the nanometric scale up to the kilometer, even under extreme conditions, and finally, of modelling in terms of composition, dimension and function.

Research

MATISSE has for objective to understand the fundamental principles governing the organization of the material to be able to develop original materials by playing on their dimensionality and feature. For the first time, the parallel between natural materials and synthetic materials is exploited in a forward-looking way by involving jointly the chemistry, the physics and the Earth sciences. This original approach will drive to a better understanding and to a control of materials in the environment (e.g. change, biodegradation, contamination), and materials for the environment (e.g. cleanup, green catalysis, management of natural resources).

Innovation

MATISSE received an exceptional support on behalf of industrial partners, what testifies from them, of a real trust in the skills of this consortium, and in its capacity to be answered the main stakes which settle in materials science today. We can quote the management of mining resources, the storage of waste, the protection against the corrosion, the soil remediation, the elaboration of materials environment-friendly

Formation

MATISSE will participate in the development of interdisciplinary and international training program at the level Master's degree and doctorate. These students and PhD students constitute a reservoir of highly considered staffs so in the academic as industrial environment.

Matisse funds full PhD and postdoc fellowships including salary and functioning. One call is open every year.

More here: www.matisse.upmc.fr





Welcome

PISACMS, Aug. 29 - Sep. 02 2016, PARIS, FRANCE

The aim of this school is to form new generations of young researchers to atomic, molecular and condensed phase computational methods, from theory to the most advanced approaches, going from the very microscopic to the macroscopic: Density Functional Theory, classical Monte-Carlo and molecular dynamics, ab initio molecular dynamics, metadynamics, linear response, lattice-Boltzmann methods, Quantum Monte-Carlo.

Our ambition is to make students aware of the fundamental principles of each methods, and then able to choose the most adapted one(s) for a given scientific problem. The broad panel of approaches we propose, both via theory lessons and computer lab "projects", is rare in the international landscape, as the best-known schools are more focalised on a particular time and length scale level of description.

This school will benefit both from the exceptional central Paris environment, and from the local concentration of young and confirmed experts-teachers in each specific theme of the school.

This school is held with the support of Labex Matisse and Sorbonne Universités.



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Organizers

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PLANS / MAPS

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Access :

-Métro:
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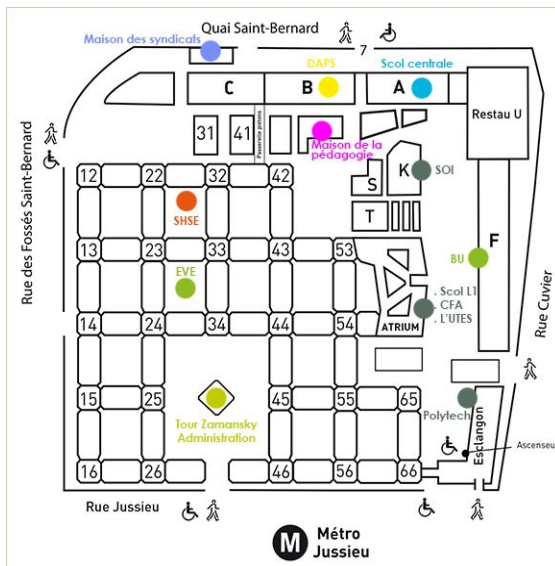
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Métro:
Ligne 7 or 10 (station: Jussieu)



Amphi 15: Morning sessions – Lunch – Coffee break – Poster sessions

22-23 – 1st floor, rooms 109-111-112-114: Computer labs

Tour Zamanski – 24th floor: Best poster prize & Closing Party



Housing and accommodation

Résidence Internationale de Paris

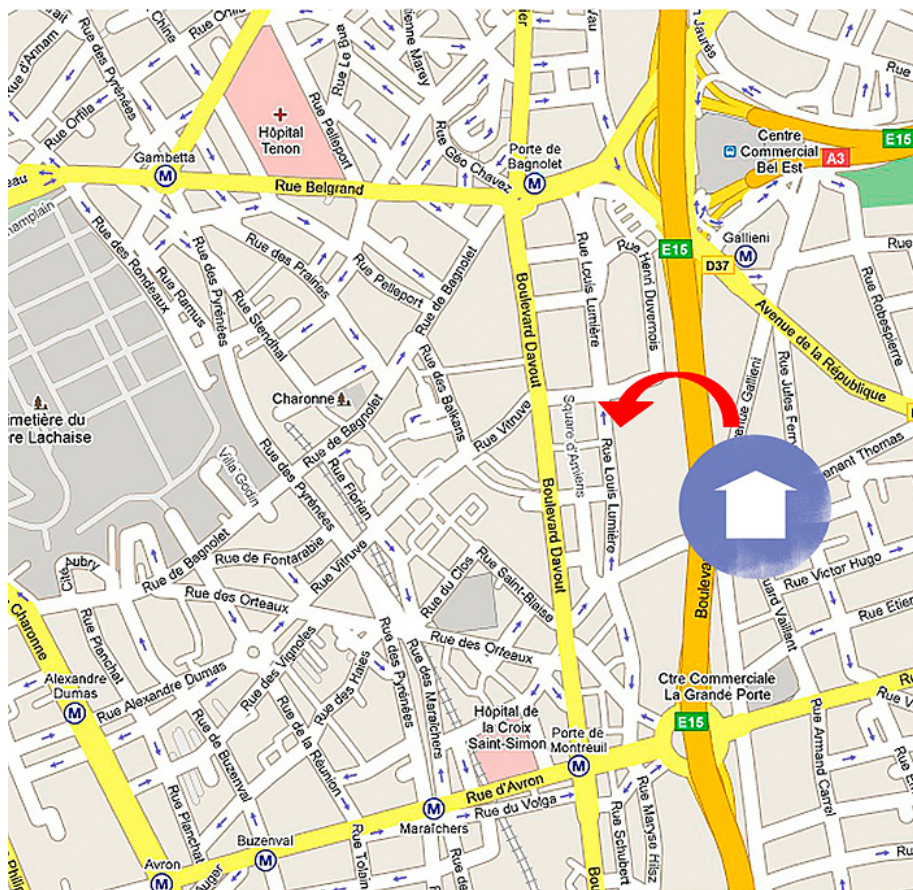
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- Bus

Lignes 57 et PC 2 (Station Vitruve).





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Program

Courses: Amphi 15

Flash presentations: Amphi 15

Computer Lab: computer rooms TBC

Lunch/coffee breaks: patio Amphi 15

	Monday 28	Tuesday 30	Wednesday 31	Thursday 1	Friday 2
9 :00	9 :00-10 :30 Molecular Dynamics simulations	9 :00-10 :30 Density Functional Theory	9 :00-10 :30 Mesoscopic methods	9 :00-10 :30 Monte-Carlo method	9 :00-10 :30 Ab initio Molecular Dynamics
10 :00	Mathieu Salanne	A. Marco Saitta	Benjamin Rotenberg	Virginie Mary	Ari P. Seitsonen
	Coffee break	Coffee break	Coffee break	Coffee break	Coffee break
11 :00	11 :00-12 :30 Free energy calculations	11 :00-12 :30 Density Functional Perturbation Theory	11 :00-12 :30 Lattice Boltzmann simulations	11 :00-12 :30 Quantum Monte-Carlo	11 :00-12 :30 Path integral simulations
12 :00	Fabio Pietrucci	Michele Lazzeri	Benjamin Rotenberg	Michele Casula	Rodolphe Vuilleumier
12 :30-13 :30	Lunch	Lunch	Lunch	Lunch	Lunch
13 :30-14 :30	Flash présentations 1	Flash présentations 2			
14 :00	14 :30-17 :30 Computer labs	14 :30-17 :30 Computer labs	14 :00-17 :30 Computer labs	14 :00-17 :30 Computer labs	14 :00-17 :30 Computer labs
15 :00					
16 :00					
17 :00					
18 :00	18 :00-20 :00 Welcome cocktail		17 :30-19 :00 Poster session 1	17 :30-19 :00 Poster session 2	18 :30-19 :30 Best poster price
19 :00					18 :00-20 :00 Closing cocktail
20 :00					



Abstracts – Courses & Computer Labs

MONDAY 28th August

Mathieu Salanne – 9h-10h30 – Amphi 15

Molecular Dynamics Simulations

Classical molecular simulations are now one of the main tools for interpreting experiments and predicting physico-chemical properties of condensed matter systems. Indeed, they provide a very useful picture of the materials at the microscopic scale. Their popularity is also due to their easy access, even for non-specialists, with a huge variety of simulation codes available.

In this lecture, we will introduce Molecular Dynamics (MD). In MD, the trajectory of the molecules is gathered by solving iteratively the Newton's equation of motion. The related algorithm, its limits and approximations, success and failures, will be detailed. The various thermodynamic and dynamic quantities that can be extracted from the simulations will also be overviewed.

Fabio Pietrucci – 11h-12h30 – Amphi 15

Free-energy calculations

Plain molecular dynamics simulations cannot sample efficiently rare events like chemical reactions, phase transitions, and many other activated processes that require overcoming free energy barriers. This lecture will present two very successful techniques employed to enhance the sampling of low-populated regions in configuration space and reconstruct free energy profiles: umbrella sampling and metadynamics. Starting from the connection between probability histograms and free energy landscapes as a function of collective variables, umbrella sampling will be discussed as a practical way to focus the computational effort in selected regions of the configuration space. The weighted histogram analysis method will then be introduced to combine together in an automatic way the statistics obtained in independent simulations. Next, the basic principles and advantages of metadynamics will be presented, together with practical considerations (by means of examples) about the choice of parameters and the convergence of the algorithm. A brief introduction to the Plumed plugin will demonstrate how to perform actual free energy calculations in the hands-on session. The last part of the lecture will





focus on topics at the core of all free energy calculation approaches: how to choose appropriate collective variables, the definition of reaction coordinates by means of committer probabilities and the nature of transition states.

Computer Lab – 14h30-17h30 – Rooms 22/23 1st floor

Search for structural minima in carbon nanotubes and clusters

Exercise 1

We will study the Stone-Wales transformation in a carbon nanotube (CNT) employing classical molecular dynamics (LAMMPS code) in combination with metadynamics (Plumed code). We will adopt simple collective variables like distances between atom pairs. The aim is setting up a simple metadynamics simulation and evaluate the effect of the different parameters (e.g., Gaussian width, height, and deposition rate) on the reconstructed free energy landscape. The tasks will include:

- choose atoms involved in collective variables from a CNT structure, using VMD
- adjust a Plumed input file to define collective variables and parameters of the simulation
- run a molecular dynamics simulation with LAMMPS + Plumed
- analyze the trajectory of collective variables to detect transitions
- plot the reconstructed free energy landscape at different times: find the “filling time”, evaluate convergence, compare barriers with literature

Exercise 2

We will explore many different geometries of Lennard-Jones clusters exploiting metadynamics in combination with topological collective variables (SPRINT). These variables characterize the network of interatomic bonds and drive the system to change topology without specifying in advance the final states, thus allowing to discover new geometries. The tasks will include:

- build initial structure of the cluster (“by hand”, or with VMD)
- adjust a Plumed input file and run a simulation with LAMMPS + Plumed
- analyze the trajectory of collective variables to spot out the metastable structures
- based on the trajectory of the potential energy, rank structures and find the global minimum



TUESDAY 29th August

A. Marco Saitta – 14h-17h – Amphi 15

Density Functional Theory

This class will establish the “common basis” of Density-Functional Theory, which will be then useful in the following of the school. After introducing the electronic density as the central quantity of the theory, the variational principle and the Hohenberg-Kohn theorem, we will follow the Kohn-Sham approach to obtain the minimization self-consistent equations, within the local-density and the generalized-gradient approximations. We will then we will briefly illustrate some practical implementation aspects, including pseudopotentials, plane-wave expansion, periodic boundary conditions, Brillouin zone sampling.

We will then consider a few basic examples of DFT calculations, from the band structure of semiconductors and metals, to charge distributions and bonding properties, to the optimization of internal coordinates and/or unit cell, to structural phase transitions, to surfaces, interfaces and low-dimensional systems. Each of these examples will feature some practical implementation techniques and exploitation of results.

Michele Lazzeri – 11h-12h30 – Amphi 15

Linear response & Density Functional Perturbation Theory

DFT is a powerful tool for the calculation of the ground-state properties of materials. In practice, however, materials are experimentally probed by measuring their response to several external perturbations. Therefore, their computation has a tremendous importance.

This can be achieved through Density Functional Perturbation Theory (DFPT), which is related to the DFT as usual perturbation theory to the Schrödinger equation. The aim of this lecture is to describe the fundamental theorems at the basis of DFPT in the case of static perturbations: the Born-Oppenheimer approximation, the Hellmann-Feynman theorem and the systematic derivation of formulas to compute n^{th} order derivatives of the total energy. Then, we will show some examples of the calculation of first- and second-order derivatives of the total energy and relate them to some selected physical examples. The lecture will be complemented by a hands-on-computer tutorial on the linear response theory applied to the study of the para-electric to ferro-electric phase transition.

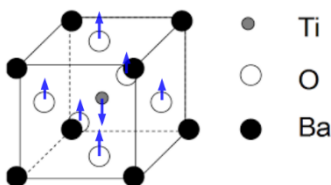


Structural Instability and Soft Phonons of cubic BaTiO₃

In this tutorial, we study the structural phase transition of the cubic, para-electric phase to the ferro-electric tetragonal phase of BaTiO₃, by using Density Functional Theory (DFT) and Density Functional Perturbation Theory (DFPT). The numerical results will be complemented by simple analytical models, when possible.

The cubic structure of BaTiO₃ is unstable with respect to a transverse optic long-wavelength mode (see Figure). This mode thus corresponds to a negative squared frequency that clearly shows up in the computed phonon dispersion $\omega(\mathbf{q})$ for $\mathbf{q}$ vectors close to zero.

The aim of this tutorial is to compute and analyze the phonon dispersion, to relate the mode that is responsible for the instability to a structure having small atomic displacements and to understand the physical origin of the instability itself. The tutorial consists of several distinct steps:



1. Determine the optimal computational parameters (cutoff energy, Brillouin-zone sampling, etc.)
2. Compute the theoretical equilibrium lattice parameter and compare it to the experimental one.
3. Run a phonon calculation at the center of the Brillouin Zone ($\mathbf{q} = 0$) and analyze the character of the soft mode by looking at the computed eigenvectors and the atom effective charges. (Be careful, this calculation could take about an hour on a small PC - launch the corresponding job and go ahead with the tutorial in the meanwhile!)
4. Run two ground-state calculations: the first for a cubic structure with atoms slightly displaced from the high-symmetry positions; the second for a tetragonal phase where atomic positions are numerically optimized. Compare the total energies of the high-symmetry cubic phase, the cubic phase with atom displaced by hand and the optimized tetragonal phase. Deduce the relative stability of those structures.



5. If time allows, compute the phonon dispersion along selected points in the Brillouin Zone. Plot the computed dispersion and compare it to experimental data.

All along this tutorial, you will use the package *Quantum Espresso*, which is based on the DFT and the DFPT. The doc is available on the website www.quantum-espresso.org.

For the ground-state properties, you will use the `pw.x` module, while the dielectric response and the vibrational modes will be computed via `phonon.x`. The analysis is done through `dynmat.x` and the suite `xcrysden`. All modules are pre-installed, no need to download! Remember also that $1 \text{ Ryd} = 13.6057 \text{ eV}$.

This tutorial is largely inspired by the Ph.D. Thesis work by Philippe Ghosez, which is available as a pdf file in the pre-installed directory. I highly recommend reading chapter 7 and some of the quoted references.

WEDNESDAY 30th August

Benjamin Rotenberg– 9h-10h30 – Amphi 15

Mesoscopic hydrodynamics

After introducing different situations involving the transport of simple and complex fluids, with applications in basic research, as well as in various industrial and environmental contexts, we will recall the most important notions in hydrodynamics. Starting from the standard macroscopic description (conservation equations, hydrodynamic regimes, etc), we will show how coming back to the microscopic and mesoscopic scales allows overcoming some fundamental and computational limitations of Direct Numerical Simulation (i.e. solving the Navier-Stokes equation with finite elements or finite volume methods).

We will consider in particular small objects (colloids, nanoparticles) suspended in a fluid or fluids confined down to the micrometer or sub-micrometer scale, and discuss the effect of thermal fluctuations, the simulation of hydrodynamic interactions, complex fluids, etc that play a key role in soft matter systems or fluids in porous media. We will introduce several mesoscopic simulation techniques allowing for the description of such systems beyond the reach of molecular simulation, including Dissipative Particle Dynamics or Stochastic Rotation Dynamics.



Benjamin Rotenberg – 11h-12h30 – Amphi 15

Lattice Boltzmann methods

In this second lecture, we will focus on a specific mesoscopic method for the simulation of simple and complex fluids. We will first introduce some basic notions of Kinetic Theory, on which the Lattice-Boltzmann (LB) method is grounded. We will sketch the derivation of the LB algorithm from continuous kinetic theory and illustrate how macroscopic hydrodynamics may emerge from a set of simple local rules. We will then extend the discussion to complex fluids such as fluid mixtures, colloidal suspensions, fluids in charged porous media, adsorption/desorption at solid/liquid interfaces. If time permits, we will also describe another related lattice-based method (Moment Propagation) that allows for the computation of time-dependent properties such as the time-dependent diffusion coefficient of tracers.

Computer Lab – 14h-17h30 – Rooms 22/23 1st floor

Transport of fluids in porous media

The students will use a Lattice-Boltzmann code developed by members of the PHENIX and PASTEUR laboratories, to investigate the transport of a simple fluid through porous media under an applied pressure gradient. Starting with the case of the Poiseuille flow in a slit pore, we will examine several aspects of LB simulations and of the hydrodynamic simulations in general: steady-state velocity profiles, linear response and permeability, transient regime and associated time scales, effect of discretization...

We will then move to the more complex case of a capillary filled with obstacles, mimicking a chromatographic column: The students will visualize the streamlines through the complex porous material and analyse the effect of the obstacles on the permeability of the column.

Finally, the driving and dispersion of tracers through the column will be considered, in the absence and in the presence of interactions between the solutes and the surface of the material.



THURSDAY 31th August

Virginie Marry – 9h-10h30 – Amphi 15

Monte-Carlo Methods

In this lecture, we will introduce an alternative way for sampling molecular configurations, the Monte-Carlo method. This method, which allowed the first evaluation of the equation of state of a fluid by Metropolis et al. as early as 1953, aims at generating a representative set of random configurations according to a specified distribution. While we lose information about the dynamics of the system, it is possible to design very efficient methods for sampling phase space. After introducing the fundamentals of the method, we will discuss the sampling of different thermodynamics ensembles and the use of biases to accelerate convergence.

Michele Casula – 11h-12h30 – Amphi 15

Quantum Monte-Carlo methods

The term “quantum Monte Carlo” refers to a large variety of methods using a Markov chain to sample the quantum partition function or the wave function. By playing with random numbers the N-body problem becomes tractable and, in some cases, exactly solvable, within the statistical noise which is inherent in any stochastic approach.

In this lecture, we will introduce the quantum Monte Carlo methods used to sample the wave function of a quantum many-body problem from first principles, i.e. by including electrons, ions, and the Coulomb interaction among all particles. The task is to find the exact ground state of any *ab initio* Hamiltonian by random numbers. In the lecture we will explain what are the different approaches to reach (or get close to) this goal, namely the variational Monte Carlo (VMC), Monte Carlo optimization, and diffusion Monte Carlo (DMC). For each, the related algorithm, its limits and approximations, success and failures, will be detailed.



Computer Lab – 14h-17h30 – Rooms 22/23 1st floor

Quantum treatment of water dimer: Potential Energy Surface

In this practical session and that of the following day, the students will investigate the quantum aspects of the hydrogen bond in a water dimer. First, the potential energy surface (PES) will be investigated using QMC.

The students will be introduced to the TurboRVB QMC code to perform QMC calculations of the water dimer. This session will be a practical guide to basis set, one-body orbitals generation, wave function ansatz, and energy minimization of the system under study. Once the setup is ready, a series of calculations will be carried out to study the QMC PES along the OH bond-stretching coordinate in the water dimer geometry. Comparison will then be made with a dissociative force-field along the same reaction coordinate.

FRIDAY 1st September

Ari P Seitsonen – 9-10h30 – Amphi 15

Ab Initio Molecular Dynamics

In this lecture, we will present the molecular dynamics approach, where the forces on the ionic cores are derived from the explicit solution of the electronic structure at each time step, or *on the fly*. In particular, we will discuss the Car-Parrinello and Born-Oppenheimer molecular dynamics approaches and their respective specificities on convergence, time-steps, equilibration, statistical averages *etc.* We concentrate on DFT-based methods, but review also the other latest developments in the field. We will then focus on their implementation in practise, with specific physical target quantities, such as vibrational dynamics and density of states, structural properties of disordered systems, thermodynamics, and diffusion, and explain how experimentally accessible observables can be evaluated, for example in neutron, infra-red, Raman and sum-frequency generation vibrational spectroscopies, NMR and EPR spectroscopies and so on. In the last part of the lecture we will present AIMD-based demonstration studies of chemical properties and structural transformations from a wide field of scientific disciplines.



Rodolphe Vuilleumier – 11h-12h30 – Amphi 15

Path-integral methods

Path-Integrals are a reformulation of quantum dynamics proposed by Feynman. In the context of statistical mechanics, the quantum canonical partition function of a system is shown to be equivalent to the classical canonical partition function of a system where every atom of the original system is represented by a closed polymer. The path-integral formulation of quantum mechanics then allows for the numerical computation of statistical quantities even for large systems in gas phase or in condensed phase.

In these lectures, we will introduce the path-integral reformulation of quantum mechanics and the classical isomorphism to a system of polymers for the computation of the canonical partition function. Numerical aspects of the sampling of the path-integral will then be discussed.

After quickly presenting some applications of the path-integral formalism to liquid water and isotope fractionation, we will discuss extension of the path-integral techniques to time-dependent quantities and, if time allows, Bose-Einstein statistics.

Computer Lab – 14h-17h30 – Rooms 22/23 1st floor

Quantum treatment of water dimer: Nuclear Quantum Effects

In this second part of this mini-project on the water dimer, the analytical force-field will be used to model the PES and the students will use the Path-Integral engine of CP2K to investigate the role of quantum effects on the structure of a water dimer. We will examine several aspects of Path-Integral simulations: initialization, convergence with number of beads, thermostating... Then we will investigate the role of quantum effects on the hydrogen bond strength and geometry by comparison with classical simulations. We will amply use visualization with VMD of the path-integral polymers to observe the effect of quantum delocalization and we will try to determine the probability that the water dimer be observed in a HO-...HOH2+ configuration with a spontaneous proton-transfer.



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Poster abstracts



Oxygen, carbon and calcium site-specific isotopic fractionation in apatite

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Apatite ($\text{Ca}_5(\text{PO}_4)_3\text{X}$, X OH, F, Cl) is the most common phosphate mineral. It frequently displays minor amounts of carbonate, occurring naturally in substitution of phosphates (B site) and channel anions (A site) in the crystal structure. Stable isotope composition of biogenic apatite, and in particular the composition ($\delta^{13}\text{C}$, $\delta^{18}\text{O}$) of structural carbonate groups, is an important geochemical marker that records environmental parameters and is widely used to infer past climates, biomineralization processes, dietary preferences and habitat of vertebrates.

The interpretation of isotopic measurements relies on the knowledge of equilibrium isotopic fractionation laws usually determined experimentally. However, out-of-equilibrium fractionation during crystal growth, slow isotope exchange rates and above all the variability of apatite crystal-chemistry are sources of uncertainties in isotopic fractionation laws established experimentally. Alternatively, equilibrium fractionation factors can be determined theoretically from the computation of vibrational properties. In this contribution, theoretical equilibrium isotopic fractionation of oxygen, carbon and calcium in hydroxyapatite and carbonate-bearing hydroxyapatite is investigated using first-principles methods based on density-functional theory. Our results revealed differences in O, C and Ca isotopic fractionation behaviour depending on the substitution site for carbonate group in apatite. Significant enrichment in heavy isotopes (^{18}O , ^{13}C) is observed in A-type carbonates relative to B-type carbonate groups in apatite (typically 7‰ for O and C fractionation at 37°C). Theoretical oxygen fractionation between B-type carbonate and phosphate ($1000 \ln \alpha(\text{CO}_3^{2-}\text{-PO}_4^{3-}) = 8.5\%$ at 37°C) is in agreement with experimental values obtained on modern and well-preserved fossil bio-apatites. In light of these theoretical results, measurements of site-specific isotopic fractionation properties would improve our understanding of isotopic records in apatites.



Atomistic computational study of multicomponent oil-brine interfaces

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^a Federal University of ABC; ^b University of São Paulo

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The atomistic study of interfacial phenomena of immiscible fluids is important for optimize enhanced oil recovery processes. Once it is well characterized, one can suggest forms to tune and control these interfaces. In this work, we have performed classical molecular dynamics of a multicomponent oil with brine interfaces to assess the interfacial tension (IFT) behavior at ambient and reservoir conditions. Our proposed oil model is a mix combination over 36 different molecules types (PNA model), combined within 60% Paraffinic (P), 25% Aromatic (A) and 15% Naphtenic (N) fractions with density of 0.8465 g/cm³. Also, we have proposed a brine model composed by NaCl and/or CaCl₂ with different salt concentrations and API brine model (8% w.t. of NaCl and 2% w.t. of CaCl₂). The interatomic potentials used were: SPCE-FH, a flexible 3-site water potential and CHARMM for the oil. We show that the IFT decreases with temperature and increases with pressure, as it is reported in literature. Also, the brine solution with both salts modifies the behavior of the IFT. This causes a more pronounced disruption of the hydrogen bond network, due the interaction of both monovalent and divalent cations. Another important trend is that the effect of the salinity was less pronounced for monovalent ions compared with divalent ones, resulting in lower IFT values for CaCl₂ in comparison with NaCl brine. The interfaces were further analyzed by determining the structural properties in the oil-brine interface, such as the density profile and the preferential order parameter for both oil and water molecules. In these interfaces, we could observe that the aromatic molecules tends to accumulate in the interface between this multicomponent oil and brine.

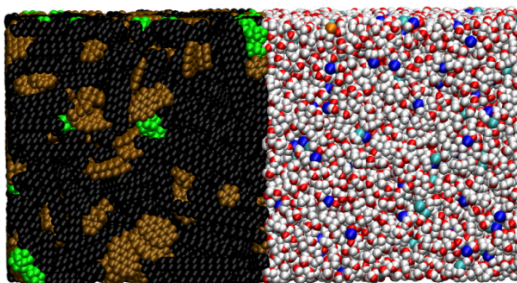


Figure 1: Snapshot of an interface between proposed oil model and an aqueous solution with NaCl and CaCl₂. Left fraction is composed by oil model and right fraction is for brine model.



Influence of terrace widths on Au(111) reconstruction[1]

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The effect of steps on the herringbone pattern appearing at the Au(111) surface is explored. Scanning tunneling microscopy investigations show that the number of alternating fcc - hcp regions decreases with the decreasing width of the terrace, in fair agreement with atomistic simulations. It is demonstrated that the steps locally release the tensile surface stresses, leading to a reorganization of the herringbone pattern.

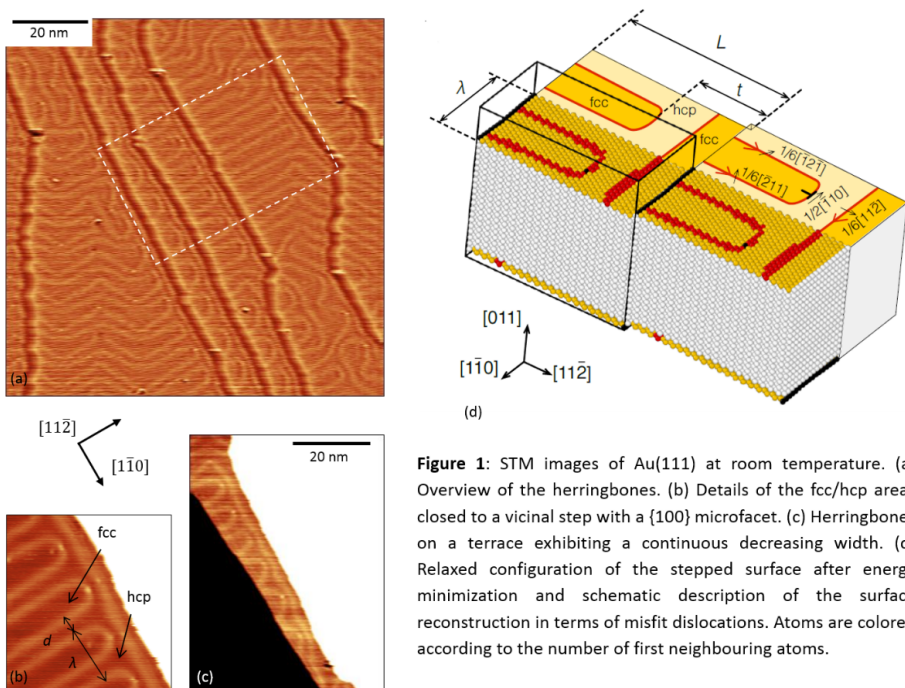


Figure 1: STM images of Au(111) at room temperature. (a) Overview of the herringbones. (b) Details of the fcc/hcp areas closed to a vicinal step with a {100} microfacet. (c) Herringbones on a terrace exhibiting a continuous decreasing width. (d) Relaxed configuration of the stepped surface after energy minimization and schematic description of the surface reconstruction in terms of misfit dislocations. Atoms are colored according to the number of first neighbouring atoms.

[1] D. Chauraud, J. Durinck, M. Drouet, L. Vernisse, J. Bonneville and C. Coupeau, Phys. Rev. B, accepted



First-principles calculation of irradiation damage in the uranium-plutonium mixed oxide (U,Pu)O₂

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The uranium-plutonium mixed oxide displays an important industrial interest, since it is used as nuclear fuel for some pressurized water reactors (PWR) in France and envisaged as reference fuel for generation IV nuclear reactors. In reactor, the collisions between fission products, having initially a high velocity, and the atoms of the fuel cause many atom displacements, leading to damage at the atomic scale. This damage, which consists initially of point defects, impacts numerous material properties, in particular, structural, electronic, atomic transport and thermodynamic properties. The knowledge of energetics and transport properties of these defects and how they influence the fuel properties is important for the improvement of the fuel modelling at the macroscopic scale.

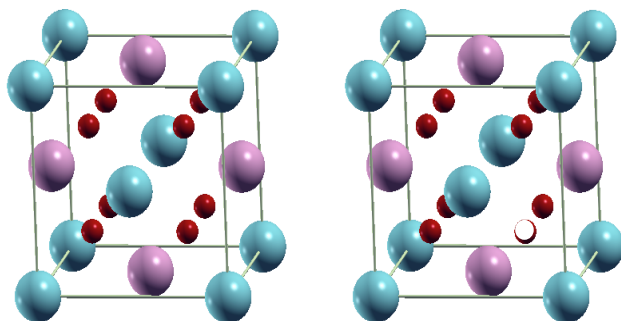


Figure 1: Point defect and its migration in the MOX fuel at the atomic scale.

Electronic structure calculations in the DFT+*U* approach [1] is a powerful tool for the modelling of the behaviour of point defects, such as oxygen or actinide vacancies and interstitials, bound Schottky defects in various charged states. In this study, we determine the point defect formation energies which allows us to find the most favourable defects formed and their impact on the structural and electronic properties of the fuel. We also study the atomic transport properties in the material to determine the migration mechanisms and activation energies. These data are needed in meso-scale models for the modelling of the evolution of the microstructure of fuel. They are also useful for the interpretation of atomic diffusion experiments.

[1] V. I. Anisimov, J. Zaanen, and O. K. Andersen, *Phys. Rev. B*, 1991, **44**, 943–954.



Surface dangling bonds and non-magnetic doping effects on Si nanowires' half-metallicity: an *ab-initio* study

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In this work, we study theoretically study the effect of B, Al, Ga, N, P and As doping on the electronic properties of Si nanowires with surface dangling bonds. The ultra-thin nanowires are modelled in the [100] crystallographic direction, with the supercell scheme. The doping is made by substituting a Si atom with a dopant atom at the centre of the cross section of the nanowire, and the dangling bonds are in mutually symmetric sites on the surface. Each nanowire spin configuration was modelled to achieve ferromagnetic, antiferromagnetic, and non-magnetic behaviours, where the optimal ground state is obtained by comparing their total energies. The electronic states due to the dangling bonds have significant spin-splitting, and the states contributed by the dopants are expected to cause a shift in the energy of the bands, allowing some bands to cross the Fermi level for one of the spin channels. The ground state magnetic behaviour is highly dependent of the dopant, for instance Al-, B- and P-doped nanowires are ferromagnetic, while Ga- and N-doped nanowires are antiferromagnetic. We hope that this investigation could enrich the knowledge about nanostructures with possible spintronics applications.



Thermal transport in high pressure phase HgTe

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Thermoelectricity is the conversion of temperature gradient into electric current and vice versa. This phenomenon is of great importance to increase energy conversion efficiency. The figure of merit (ZT) for thermoelectric materials is the quantity that reflects how efficient a material is. A quantity that is inversely proportional to the thermal conductivity. Which means that for a better thermoelectric material, low thermal conductivity is to be achieved. In the literature, the thermal conductivity of HgTe (mercury telluride) was calculated for the zincblende (low pressure) and cinnabar (high pressure) phases by solving the Boltzmann transport equation using second- and third-order force constants calculated from Density Functional Theory. The calculations for the zincblende phase have shown an almost five times higher lattice thermal conductivity than the experimental values [1]. To better understand the discrepancy and the phonon scattering processes in both Zincblende and cinnabar phases, our recently proposed residual analysis method [2] is used and we find an unusual forth-order phonon anharmonicity. Time domain normal mode analysis and other methods were conducted to achieve more accurate values for thermal conductivity and gain better understanding the abnormal phonon transport in thermoelectric material HgTe.

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[2] S Y Yue, X Zhang, G Qin, S R Phillpot, and M Hu, *Physical Review B* (2017) **95**, 195203



Tailoring nucleation and stress magnitude during surfactant-mediated metallic film growth

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Polycrystalline metallic films growing in a Volmer-Weber mode, as typically encountered during condensation of metal vapors on insulating substrates, are technologically important in a variety of applications, such as catalysis or plasmonics for which nanoparticle synthesis is a requisite. The nucleation, coalescence and continuous film growth stages of high-mobility metals are associated with a compressive-tensile-compressive behavior, as evidenced using real-time stress diagnostic.

As the lateral grain size is usually set-in during the coalescence stage, controlling the nucleation conditions is of paramount importance for tuning the films properties as well as stress levels. This can be achieved by changing the substrate temperature or deposition flux.

Here we employ Ge surfactant to vary the nucleation conditions during sputter-deposition of Cu films, and use a combination of in situ and real-time techniques to monitor the growth. We identify the percolation thickness from electrical resistivity measurements, while the onset of film continuity is determined from the maximum tensile force using a multiple beam optical stress sensor. Complementary analyses using atomic force and transmission electron microscopy are employed to obtain information on the growth morphology and grain size.



Chemisorption of Hydroxides on Carbon and Boron Nitride Nanomaterials from *Ab Initio* Calculations

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Recent nanofluidic experiments revealed strongly different surface charge measurements for boron-nitride (BN) and carbon nanotubes when in contact with saline and alkaline water.^{1,2} These observations contrast with the similar reactivity of a graphene layer and its BN counterpart, using density functional theory (DFT) framework, for intact and dissociative adsorption of gaseous water molecules. We investigate, by DFT in implicit water, single and multiple adsorption of anionic hydroxide on single layers.³ A differential adsorption strength is found in vacuum for the first ionic adsorption on the two materials - chemisorbed on BN while physisorbed on graphene. The effect of implicit solvation reduces all adsorption values, resulting in a favorable (nonfavorable) adsorption on BN (graphene). We also calculate a $pK_a \simeq 6$ for BN in water, in good agreement with experiments. Comparatively, the unfavorable results for graphene in water echo the weaker surface charge measurements but point to an alternative scenario.

Various carbon and boron nitride allotropes are currently tested to probe possible effects of stacking, chirality and curvature. The adsorption of alternative ions are also investigated to determine the charging mechanism of the carbon surface.

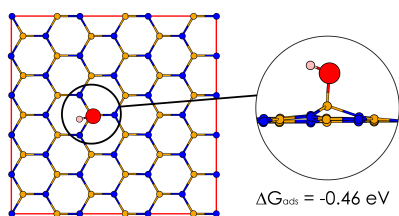


Figure 1: DFT adsorption structure for hexagonal boron nitride and associated Gibbs free energy of adsorption in implicit water at room temperature and pressure

- [1] Siria A, Poncharal P, Biance A-L, Fulcran R, Blasé X, Purcell S T, Bocquet L, *Nature*, 2013, **494**, 455-458.
- [2] Secchi E, Niguès A, Jubin L, Siria A, Bocquet L, *Physical Review Letters*, 2016, **116**, 154501.
- [3] Grosjean B, Péan C, Siria A, Bocquet L, Vuilleumier R, Bocquet M-L, *Journal of Physical Chemistry Letters*, 2016, **7**, 4695-4700.



kMC simulations of transition metal nitride thin film growth at Oblique Angle Deposition process

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During the past years Oblique Angle Deposition (OAD) has received an increasing popularity as it has allowed tailoring unique film microstructure. It's a way to architecture the growing layer and then control film properties, as texture, porosity, and anisotropy. However, predicting the film microstructure still remains a challenging problem, as the influence of the process parameters is not well understood. If OAD has been largely investigated for the case of thermal evaporation, less data are available for sputter-deposited films. In particular, the role of temperature, working pressure, angular and energy distribution of the impinging particles on the resulting microstructural attributes, such as the tilt angle of the column with respect to the normal of substrate but also the crystal structure, texture, grain size and growth defects, remains to be systematically explored. For titanium nitride (TiN) films synthesized by reactive magnetron-sputter deposition, the resulting film morphology is usually columnar, even at normal incidence. Investigations on TiN film morphologies obtained at OAD remain scarce. In this context, predictive design can be achieved, by using multi-scale computational approaches based on kinetic Monte Carlo (kMC) algorithm to mimic real deposition conditions, and gain insights on understanding for competitive columnar growth and texture evolution for TiN based films. In this framework, a kMC code which takes into account surface diffusion events of metal (Ti) and nitrogen particles, on a rigid 3D lattice, has been developed [1]. In order to reproduce experimental findings of competitive columnar growth an accurate model of the adatom diffusion anisotropy on (111), (100) or (110) surfaces coming from density functional theory (DFT) calculations [2] has been also implemented. The present work gives some simulation results of the developed code for reactive magnetron sputter-deposition of TiN at OAD. The influence of the most important experimental parameters (substrate temperature, deposition pressure, incident angle and deposited energy of some incoming particles) will be discussed regarding the resulting thin film morphology (porosity, surface roughness, column tilt angle, column width, layer density). The results will be compared with some experimental measurements on magnetron sputter-deposited TiN films at oblique angles of 5°, 35°, 65°, 75° and 85°, in the 0.3-0.5 Pa and 300-750 K range, as well as kMC results obtained from Simul3D software.

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Phase transformation of CO₂ under extreme conditions

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CO₂ is largely present in the earth's mantle and is one of the main constituents of the volcanic emission. In the recent years several phases have been discovered and investigated [1,2] in the geological conditions of the upper mantle of Earth (5-80 GPa, 100-2000 K) but many questions the transition mechanisms between those phases still remain, meanwhile a good knowledge of their property and transformations is key to understand the effect of CO₂ on the geological properties of the mantle. In this work we aim at getting a better understanding of those transitions using both ab-initio (DFT) and classical molecular dynamics coupled with enhanced sampling techniques such as metadynamics to sample the free energy landscape of the system.

[1] Datchi, F., Mallick, B., Salamat, A., & Ninet, S., *Physical review letters*, 2012, **108**(12), 125701.

[2] Boates, B., Teweldeberhan, A. M., & Bonev, *Proceedings of the National Academy of Sciences*, 2012, **109**(37), 14808-14812.



Is the step interaction attractive?

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We have investigated the possible attractive interaction between steps on the surface $\beta 2 \times 4$ reconstructed GaAs (001) which has been observed experimentally [1], [2] on other surfaces. Attractive interactions between surface steps are believed to not exist [3].

In order to do this, we have considered two vicinal surfaces on the $\beta 2 \times 4$ reconstructed GaAs (001) surface misoriented towards the (111) surface, with the step termination named as Aa and Ab.

The calculations have been carried out using the open source suite QUANTUM ESPRESSO, which implements an ab-initio approach based on the density functional theory. We have calculated the step energy and the step-step interaction fitting the results to the generally used expression for the surface energies of unstrained surfaces. We have found an attractive step-step interaction, in contrast with the Marchenko-Parshin model [3], which predicts a repulsive attraction between like-oriented steps.

We have investigated the different contributions to the surface energy and identified the main attractive interaction term.

Furthermore, we have isolated the strain energy contribution to the step interaction, which is very small for this kind of reconstructed surfaces.

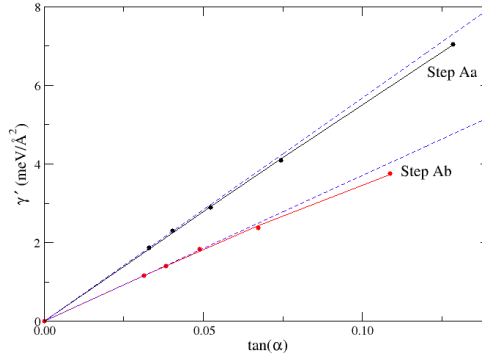


Figure 1: Surface energy versus miscut angle $\tan(\alpha)$, the black and the red points are the surface energies for step Aa and step Ab, respectively. The curve is the fit of the values. The straight lines are the trend for isolated steps.

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A multi-scale computational study unveils the molecular mechanism responsible for color expression in anthocyanins

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The use of pigments and dyes is well spread in the history of human beings. This includes traditional color applications in the arts, as well as in the industry, e.g. in the cosmetic, textile, and food sectors. Several artificial pigments have been recently suspected to have adverse effects on the health of consumers, leading to a growing and urgent interest in the use of natural colorants to reproduce all range of colors in the food industry. Anthocyanins are among the most common pigments, responsible for most colors in the red-purple-blue gamut found in nature, depending on the physical and chemical features of the environment (such as, e.g. temperature, acidity, copigmentation). In this poster I will report on a theoretical study, based on a newly established multiscale simulation protocol, which has allowed us to unravel the electronic and structural mechanisms responsible for color expression in a few representative molecules of the anthocyanin family. Our protocol relies on (i) enhanced-sampling classical molecular dynamics (MD) simulations to identify the most representative conformers of the molecules in explicit solvent [1]; (ii) *ab initio* MD of the most representative conformers in explicit solvent (see figure) and (iii) time-dependent density-functional theory (TDDFT) on selected frames sampled from the *ab initio* MD of the distinct conformers, performed using a newly developed implicit-solvent model [2]. The spectra are, finally, calculated as weighted average over the different conformers sampled.

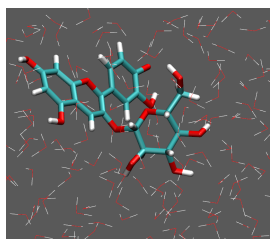


Figure: Cyanin in water solution

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[2] I. Timrov and e.c *J. Chem. Theory Comput.* 2016, **12**, 4423-4429



Thermal transport in layered and 2D materials within the relaxons picture

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Layered and 2D materials are promising platforms for next generation electronic devices [1]. In general, the bottleneck for miniaturization of electronic devices is overheating [2], which is related to the thermal transport properties of the material. 2D materials feature a thermal conductivity larger than their 3D layered parents and novel thermal transport regimes, which are at the origin of exotic phenomena such as the breakdown of Fourier's law and size dependent thermal conductivity [3].

The aim of this project is to understand the differing thermal properties of 3D layered materials and 2D monolayers obtained from them. Recently it has been shown [4] that the microscopic mechanism underlying thermal transport can be explained in terms of a gas of collective phonon excitations, called "relaxons" and defined as the eigenvectors of the scattering matrix appearing in the phonon Boltzmann equation (BTE).

We solved exactly the BTE in the relaxons picture for graphite, graphene, MoS₂ bulk and monolayer. Our preliminary results show that thermal properties of 2D monolayers and their parent 3D layered materials are very different: the former show an higher thermal conductivity than the latter and in 2D the heat is carried by fewer relaxons than in 3D. In the future we would like to understand and formalize the novel physics exposed by the relaxons formalism and collaborate with experimentalists to measure relaxon lifetimes and mean free paths.

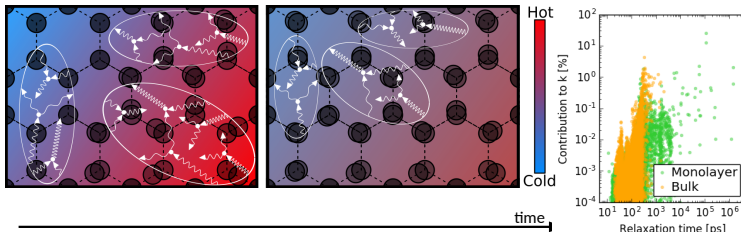


Figure 1: From left to right: pictorial representation of relaxons, i.e. a linear combinations of interacting phonon wavepackets which are created when the material is driven out of equilibrium through a thermal excitation. In the center it is shown that each relaxon evolves independently from the others and it moves inside the material carrying heat and disappearing at a rate determined by its relaxation time. On the right, comparison of relaxon lifetimes vs contribution to the thermal conductivity in MoS₂ bulk (3D, orange) and monolayer (2D, green). In 3D heat is carried by a large number of relaxons having lifetimes around 200 ps. The 2D thermal conductivity has a dominant contribution coming from two relaxons having a much longer lifetime, around 1 ns.

- [1] Q. H. Wang, K. Kalantar-Zadeh, A. Kis, J. N. Coleman, and M. S. Strano, *Nature Nanotechnology*, **2012**, 7(11), 699-712.
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Molecular dynamics study of thermal and dynamical properties of Ag_2Te

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Thermoelectric (TE) materials, which can directly and reversibly convert heat to electricity, provide a promising waste-heat-recovery solution to improve energy sustainability. Despite vast theoretical and experimental efforts, physical mechanisms of thermal transport in some promising high efficiency TE materials, e.g. ordered-disordered systems, have still not been well understood. As one typical ionic crystal with ordered-disordered phase at high temperature, Ag_2Te presents an extremely low thermal conductivity. In this study, we calculated the thermal conductivities and their contributing factors of Ag_2Te in the high temperature range (500K – 1000K) using molecular dynamics method to understand the underlying heat transfer mechanisms. We also investigated the diffusion properties of constituent atoms of Ag_2Te and illustrate the unique lattice dynamics feature of highly disordered cations with liquid-like mobility around the anion sublattice. We also used the equilibrium ab initio molecular dynamics (AIMD) in determining dynamical properties of Ag_2Te for comparison and validation. Our results show an anomalous evolution of thermal conductivities with temperature of Ag_2Te and indicate how the Ag ions mobility affects the thermal transfer properties. This study is expected to advance the understanding of thermal transport in broad ordered-disordered materials and improve the TE performance by reducing the lattice thermal conductivity.

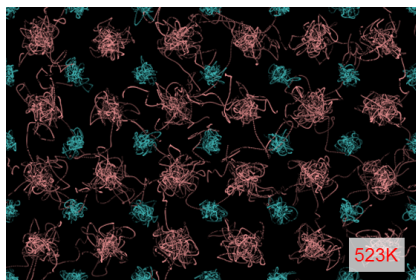


Figure 1: Atomistic trajectories of Ag_2Te for 10 ps at 523 K.

