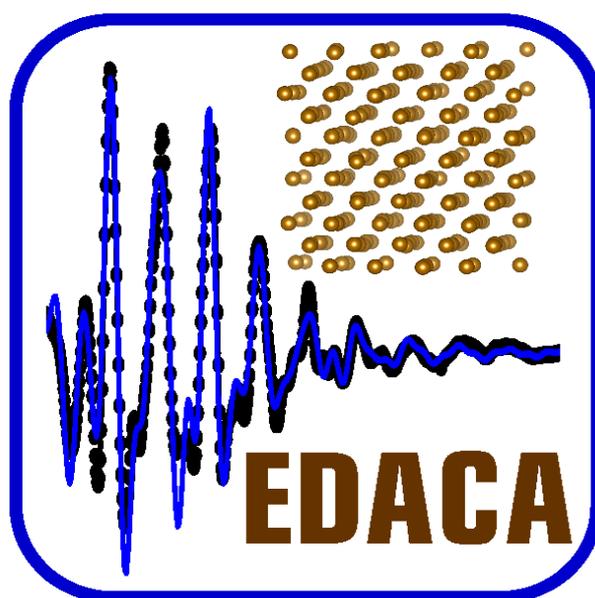


Alexei Kuzmin

# EDACA



## User's Manual

Version 3.0



INSTITUTE OF SOLID STATE PHYSICS  
UNIVERSITY OF LATVIA

# Preface

X-ray absorption spectroscopy (XAS) at synchrotron radiation sources is a structural tool providing information on the local atomic and electronic structure around an atom of a particular type. Today XAS is successfully applied to a study of crystalline, nanocrystalline and disordered solids, liquids and gases in a wide range of external conditions defined by temperature, pressure, etc. The size of the region, probed by XAS, depends on the degree of thermal and static disorder present in a material and is limited by the mean-free path of the excited photoelectron. Typically the information reach region extends up to 3-10 Å around the absorbing atom.

An advantage of the XAS method is its sensitivity to many-atom distribution functions, giving rise to multiple-scattering (MS) contributions, and correlation effects in atom dynamics. Note that an accurate account of both effects is still challenging.

The time scale ( $\approx 10^{-15}$ - $10^{-16}$  s) of the X-ray absorption process is much shorter than the characteristic time ( $\approx 10^{-13}$ - $10^{-14}$  s) of thermal vibrations. Therefore, the atoms may be considered frozen at their instantaneous positions during a single photoabsorption process, and the total experimentally measured X-ray absorption spectrum corresponds to the configurational average of all atomic positions over the time of the experiment. This situation can be straightforwardly modelled by combining the molecular dynamics (MD) simulation with the extended X-ray absorption fine structure (EXAFS) calculations, known as the MD-EXAFS approach.

Finally, the agreement between the experimental and configuration-averaged EXAFS spectra can be used to validate the accuracy of the interatomic potential (force-field) models employed in the MD simulations.

Similarly to the EXAFS case, the full-multiple-scattering (FMS) calculations can be performed for each atomic configuration and the configuration-averaged XANES spectrum can be compared with the experimental one.

In this manual, the software code **EDACA** (from “EXAFS **d**ata **a**nalysis using **c**onfigurational **a**verage”) implementing both MD-EXAFS and MD-XANES methods is described.

## The code can be downloaded at

<http://www.dragon.lv/edaca>

## References to EDACA code:

- A. Kuzmin, R.A. Evarestov, Quantum mechanics-molecular dynamics approach to the interpretation of X-ray absorption spectra, *J. Phys.: Condens. Matter* **21** (2009) 055401 (6 pp).  
DOI: 10.1088/0953-8984/21/5/055401
- A. Kuzmin, A. Anspoks, A. Kalinko, J. Timoshenko, The use of x-ray absorption spectra for validation of classical force-field models, *Z. Phys. Chem.* **230** (2016) 537-549.  
DOI: 10.1515/zpch-2015-0664

# MD-EXAFS & MD-XANES methods

The general scheme of the MD-EXAFS & MD-XANES methods is shown in Fig. 1 [1]. First, the structural model of the system should be constructed, taking into account its periodicity, the presence of defects as well as size and shape in the case of nanocrystals and clusters. Second, the suitable interatomic potential (force-field) model should be selected, and its parameters should be provided. Next, the molecular dynamics (MD) simulation at the required temperature and pressure is used to generate a set of instantaneous atomic configurations (“snapshots” of the material structure), and a set of EXAFS or XANES signals is calculated for each of the atomic configurations within ab initio multiple-scattering (MS) or full-multiple-scattering (FMS) approach [2-6]. Finally, the configuration-averaged EXAFS or XANES spectrum is calculated and directly compared with the experimental one. Note that in such an approach both thermal and static disorder effects are naturally accounted for within theoretical EXAFS/XANES.

Note that in the case of classical MD, the effect of zero-point vibrations is neglected, however, it becomes progressively important in the low-temperature limit  $T \rightarrow 0$  K [7]. The inclusion of quantum effects can be realized using the path-integral implementation of MD (PIMD) [8-10].

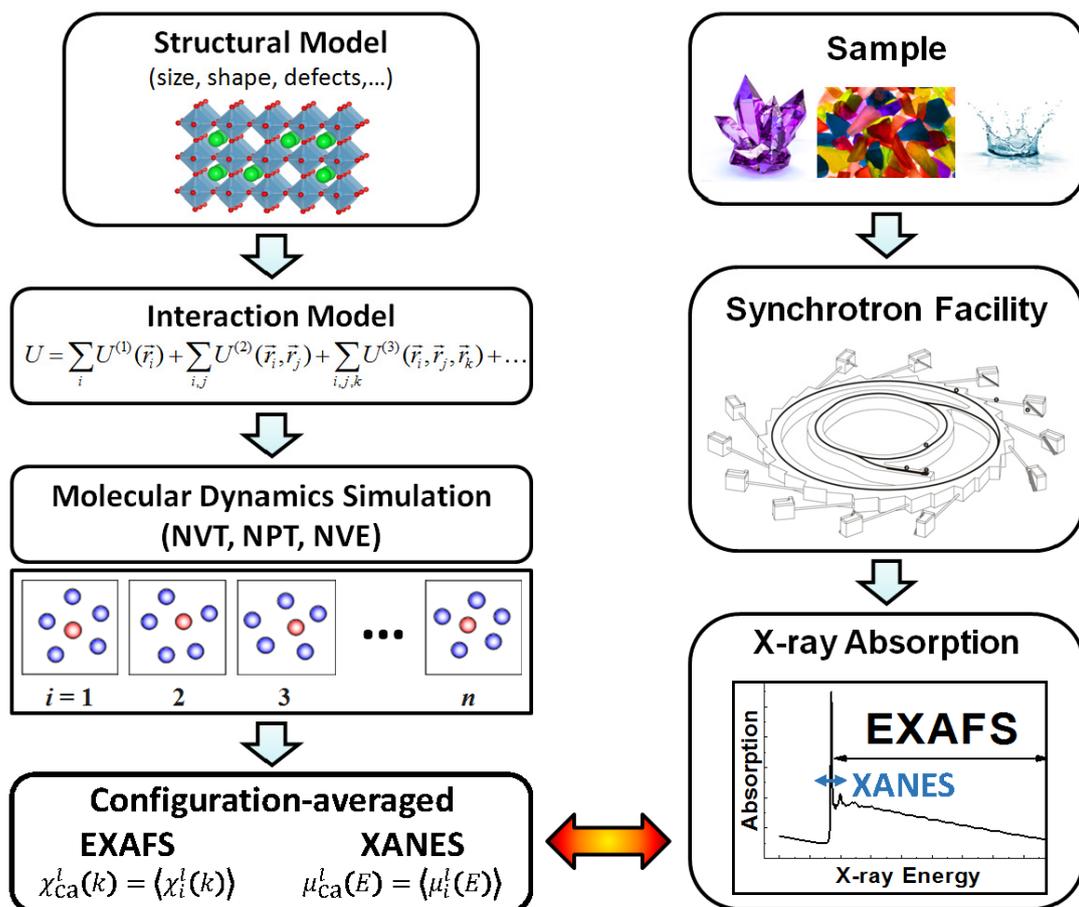


Fig. 1. Scheme of the MD-EXAFS & MD-XANES methods. See text for details.

# Basics of classical Molecular Dynamics

Molecular dynamics (MD) is a technique to simulate the motion of atoms under predefined conditions, such as **temperature** (T), **pressure** (P), etc [11-13]. Nowadays it allows one to examine the atomic structure and dynamics of materials at the atomic level and nanoscale on time scales up to nanoseconds and more [14]. Moreover, large-scale MD simulations, involving millions of atoms, become feasible nowadays [15, 16].

In the **NVT** ensemble, also called the *canonical* ensemble, the number (N) of particles (atoms) and the supercell size (V) are fixed, whereas the temperature (T) is controlled using a surrounding virtual heat bath (**thermostat**) [17, 18].

In the **NPT** ensemble, also called the *isothermal-isobaric* ensemble, the number (N) of particles (atoms) is fixed, whereas the temperature (T) and pressure (P) are controlled using a **thermostat** and a **barostat** [17-19].

During classical MD simulation, **Newton's equations** of motion are numerically integrated for a set of atoms starting from a given **initial configuration**. This is performed via numerical integration by discretizing time into small intervals called the **time step** ( $\Delta t$ ). The time step must be smaller than the fastest vibrational frequency in the system, typically in the order of 1 femtosecond ( $\approx 10^{-15}$  s).

The interactions between the atoms, i.e. the interatomic forces, can be calculated based on **empirical** or **machine-learning potentials** or **first-principles theory** (*ab initio*). Several databases of empirical interatomic potentials (force fields) are available at

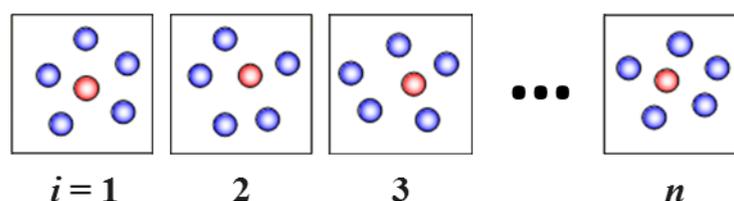
<https://gulp.curtin.edu.au/gulp/models.cfm>

<https://www.ucl.ac.uk/klmc/Potentials/>

<https://www.ctcms.nist.gov/potentials/>

<https://openkim.org/>

The interatomic forces determine the acceleration of the atoms, allowing the positions and velocities to propagate towards the next time step. Repeating this procedure many times yields a **series of snapshots** (Fig. 2), describing the trajectory of the system in phase space, which can be analyzed to extract the desired properties, including EXAFS/XANES.



**Fig. 2.** A series of atomic configuration snapshots.

Many MD codes have been developed in the past. Four of them are known to be compatible with the EDACA code:

**GULP** - the General Utility Lattice Program.

<http://gulp.curtin.edu.au/gulp/>

**LAMMPS** - a Large-scale Atomic/Molecular Massively Parallel Simulator.

<http://lammmps.sandia.gov/>

**DL\_POLY** - a general-purpose serial and parallel molecular dynamics simulation package.

[https://www.scd.stfc.ac.uk/Pages/DL\\_POLY.aspx](https://www.scd.stfc.ac.uk/Pages/DL_POLY.aspx)

**CP2K** - a program to perform atomistic and molecular simulations of solid state, liquid, molecular, and biological systems.

<https://www.cp2k.org/>

All these codes can save the MD trajectory as a standard \*.XYZ ASCII file with a specific structure (see [https://en.wikipedia.org/wiki/XYZ\\_file\\_format](https://en.wikipedia.org/wiki/XYZ_file_format) for details):

```
<number of atoms>
comment line
<element> <X> <Y> <Z>
...
<number of atoms>
comment line
<element> <X> <Y> <Z>
...
<number of atoms>
comment line
<element> <X> <Y> <Z>
...
```

One can also recommend controlling the results of the MD simulation using a visualization code. Two such codes are known to be able to read \*.XYZ ASCII file:

**VESTA** – a 3D visualization program for structural models, volumetric data such as electron/nuclear densities, and crystal morphologies.

<http://jp-minerals.org/vesta/en/>

**OVITO** – a scientific visualization and analysis software for atomistic simulation data.

<https://ovito.org/>

**VMD** – a molecular visualization program for displaying, animating, and analyzing large biomolecular systems using 3-D graphics and built-in scripting.

<http://www.ks.uiuc.edu/Research/vmd/>

The accuracy of configuration-averaged EXAFS calculations is limited mainly by the accuracy of the MD simulations. The choice of interatomic potential (force-field) model, equilibration time, proper averaging (number of configurations & time step) and the size of the simulation box is crucial.

The size of the simulation box must be large enough to avoid boundary condition artefacts such as, for example, artificial correlations. Therefore, the simulation box size must be at least twice larger than the radius of the most distant coordination shell required for the analysis. The number of atomic configurations must be larger than 100: the recommended value is about 1000 or more. The recommended time step is 0.5 fs, however, it should be modified depending on the system and conditions (T, P).

## EXAFS engine

EDACA code uses ab initio real-space multiple-scattering **FEFF8x/9x** code [2-4] to calculate EXAFS for each atomic configuration (“snapshot”). A free version of the FEFF8 code, FEFF8-Lite, restricted to EXAFS calculations can be obtained from

<https://feff.phys.washington.edu/feffproject-feff-download.html>.

**One must be familiar with the use of the FEFF code before attempting to use the EDACA code!**

Note that since the calculation of cluster potential is a time-consuming part of the EXAFS calculation, one can perform it only once at the beginning for some average atomic structure.

The FEFF commands relevant to the MD-EXAFS simulations are:

- Multiple-scattering series truncation:  
**NLEG** = 8 default
- Criteria to limit the number of paths:  
**CRITERIA** 4.0 2.5 default critcw=4.0% critpw=2.5%  
**CRITERIA** 0 0 use all paths (cw and pw criteria turned off - **recommended**)
- A “configuration” average over the spectra of multiple absorbing atoms (for nanoparticles):  
**CFAVERAGE** iphabs nabs rclabs  
**iphabs** potential index for the type of absorbing atoms over which to make the configuration average  
**nabs** the configuration average is made over the first nabs absorbers in the `feff.inp` file of type iphabs  
**rclabs** radius to make a small atom list from a bigger one allowed in `feff.inp`

**Please consult the FEFF documentation for any questions regarding the proper use of commands.**

# XANES engines

EDACA code can use two ab initio real-space full-multiple-scattering codes

- **FEFF8x/9x** code [2-4] and
- **FDMNES** [5,6]

to calculate XANES for each atomic configuration (“snapshot”).

The FEFF8x/FEFF9x code can be obtained from

<https://feff.phys.washington.edu/feffproject-feff-download.html>.

The FDMNES code is available from

<https://fdmnes.neel.cnrs.fr/>.

**One must be familiar with the use of the FEFF or FDMNES codes for XANES calculations before attempting to use the EDACA code!**

## **FEFF case**

Note that since the calculation of cluster potential is a time-consuming part of the calculation, one can perform it only once at the beginning for some average atomic structure.

The minimum set of FEFF commands relevant to the MD-XANES simulations are:

- XANES calculations up to  $k_{\max}=4.0 \text{ \AA}^{-1}$  with the steps  $\Delta k=0.05 \text{ \AA}^{-1}$  and  $\Delta E=0.2 \text{ eV}$ :

**XANES** 4.0 0.05 0.2

- Compute full multiple scattering within a sphere of radius  $8.0 \text{ \AA}$ :

**FMS** 8.0

- A “configuration” average over the spectra of multiple absorbing atoms (for nanoparticles):

**CFAVERAGE** iphabs nabs rclabs

**iphabs** potential index for the type of absorbing atoms over which to make the configuration average

**nabs** the configuration average is made over the first nabs absorbers in the ‘feff.inp’ file of type iphabs

**rclabs** radius to make a small atom list from a bigger one allowed in ‘feff.inp’

**Please consult the FEFF documentation for any questions regarding the proper use of commands.**

## ***FDMNES case***

FDMNES code calculates XANES using two techniques: (i) the Green function formalism (multiple-scattering) on a muffin-tin potential and (ii) the Finite Difference Method (FDM) to solve the Schrödinger equation. The first approach is much faster but less accurate. Besides, the calculations can be performed using self-consistent (SCF) and non-self-consistent potential as well as different multipolar expansions (dipole, quadrupole, etc). There are many advanced possibilities implemented in the FDMNES code, so please consult the documentation.

The minimum set of FDMNES commands relevant to the MD-XANES simulations are:

- The energy range (in eV) relative to the Fermi level ( $E_{\min}$ , step,  $E_{\max}$ , ...):

**Range**

-20.0 1.0 -10.0 0.2 0.0 0.2 15.0 0.5 20.0 1.0 40.0 2.0 100.0

- The Green function formalism (multiple-scattering) on a muffin-tin potential:

**Green**

- The absorption edge (K, L1, L2, L3, M1, ...):

**Edge**

K

- Cluster radius for XANES calculations:

**Radius**

8.0

- Cluster structure followed by the unit cell parameters (the atom positions will be automatically added from XYZ file below these lines) :

**Molecule**

1.0 1.0 1.0 90. 90. 90. = a, b, c, alpha, beta, gamma

Please consult the FDMNES documentation for any questions regarding the proper use of commands.

## Configuration averaged EXAFS/XANES calculations

The EDACA code is composed of three programs **edamd.exe**, **edaca.exe** and **stdev.exe**. To run EDACA code, one can use the **runedaca.bat** file, which contains the following lines:

```
edamd.exe Fe_bcc_300K_MD.xyz 0 1 8.0 Fe 0  
edaca.exe  
stdev.exe
```

As an alternative, the three programs can be run separately without employing the **runedaca.bat** file.

The **edamd.exe** code requires six or more parameters to be specified:

```
edamd.exe filename_xyz SkipFirst Skip Rmax atom1 absorber_number atom2 atom3 ...
```

*filename\_xyz* = The name of XYZ file with atomic coordinates from GULP (DL\_POLY, LAMMPS, ...)

*SkipFirst* = A number of configurations to skip from the beginning, default *SkipFirst*=0

*Skip* = A number of configurations to skip (e.g., skip=0 means to read all configurations, *Skip*=1 means to read each second configuration).

*Rmax* = The radius (in Å) of the small cluster around the absorber used in the EDACA calculations and generated from a large XYZ file. *Rmax* must be larger than the maximum distance (RPATH) specified in the feff.dat file.

*atom1* the absorber

*absorber\_number* = 0 default automatic mode: the absorber located at the MD box centre,

>0 equals the absorber number in the list of atoms,

<0 then *|Absorber\_number|* is equal to a number of possible absorbers from the beginning of the XYZ file: this option is required to calculate configuration averaged EXAFS/XANES for a part of atoms, e.g. in the central box of MD or RMC simulation.

*atomX* = next atom in the compound, those potential is mentioned in the pot.dat file.

For example:

```
edamd.exe CuMD.xyz 0 0 10.0 Cu 0           for Cu metal  
edamd.exe ReO3MD.xyz 0 0 10.0 Re 0 0      for ReO3 crystal  
edamd.exe SrTiO3MD.xyz 0 0 10.0 Ti 0 Sr 0  for SrTiO3 crystal
```

The order of elements in the command line is important and should correspond to that in the pot.dat file (see below), the first atom is always the absorber.

After running **edamd.exe**, XYZ file produced by the MD simulation will be split into a set of smaller XYZ files. Each small XYZ file corresponds to the individual atomic configuration and is centred at the proper absorbing atom ('0' number) surrounded by other atoms ('1', '2', ...) located within a sphere with the radius *Rmax*. The full list of small XYZ file names is written in a **conf.dat** file. Partial pair distribution functions are also calculated and are written in **g2\_tot.txt**, **g2\_\*.txt** files.

Examples of the **conf.dat** files created automatically by the edamd.exe code for SrTiO<sub>3</sub> and Cu:

```
SrTiO3_555_TMB_300K_1.xyz
SrTiO3_555_TMB_300K_2.xyz
SrTiO3_555_TMB_300K_3.xyz
SrTiO3_555_TMB_300K_4.xyz
....
SrTiO3_555_TMB_300K_2000.xyz
```

```
Cu_fcc_300K_MD_11.xyz
Cu_fcc_300K_MD_22.xyz
Cu_fcc_300K_MD_33.xyz
Cu_fcc_300K_MD_44.xyz
....
Cu_fcc_300K_MD_39996.xyz
```

The **edaca.exe** code uses the results produced by the edamd.exe code plus a number of additional files, which should be located in the same directory.

A set of files required by the edaca.exe includes:

- 1) a set of small XYZ files and **conf.dat** file produced by edamd.exe.
- 2) **pot.dat** a file in ASCII format which describes correspondence between elements (potentials) in the MD simulation and FEFF/FDMNES calculation. The order of elements (potentials) is important and should be checked by the user!

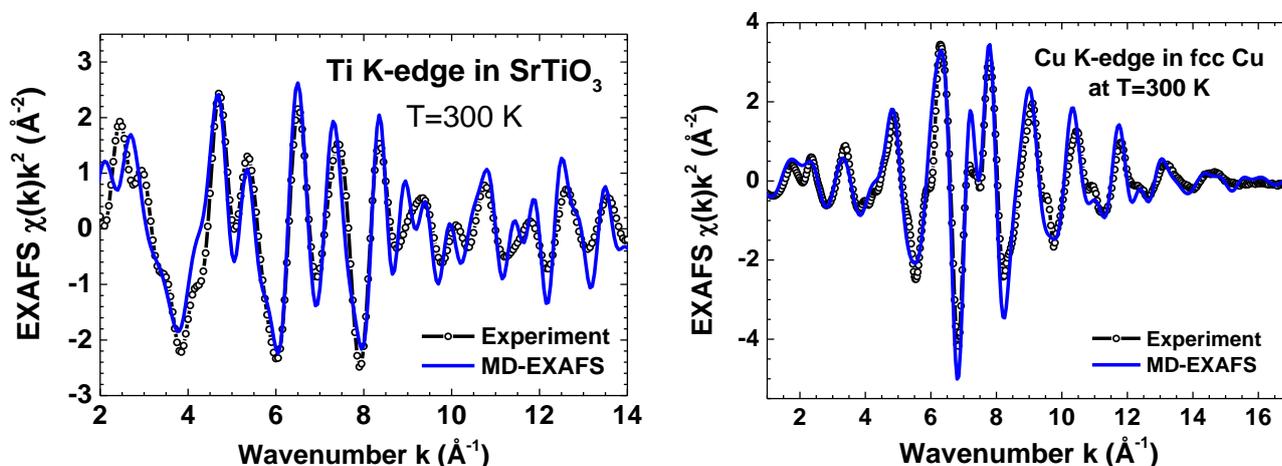
In the case of the FEFF code:

- 3) **feff.exe** an executable of the FEFF8x or FEFF9x code.
- 4) **feff.dat** an input file in ASCII format for the FEFF8x or FEFF9x code with the \*.dat extension: it can be produced from the feff.inp file simply by deleting all atomic coordinates after ATOM command.
- 5) **\*.bin** and other files required by the FEFF code (output files after FEFF8 & FEFF9 calculation for static configuration), if cluster potential is supplied by the user (recommended).

In the case of the FDMNES code:

- 3) **fdmnes.exe** an executable of the FDMNES code.
- 4) **fdmnes.dat** an input file in ASCII format for the FDMNES code with the \*.dat extension: it can be produced from the FDMNES input file simply by deleting all atomic coordinates. See the example below.

The edaca.exe code calculates EXAFS or XANES spectrum for each XYZ file specified in the conf.dat file. These spectra are saved under the names xt.001, xt.002, ... or mu.001, mu.002, .... The main result is saved under the name **xt\_tot.txt** or **mu\_tot.txt** and contains the configuration-averaged EXAFS  $\chi_{ca}^l(k)$  (Fig. 3) or XANES  $\mu_{ca}^l(E)$ .



**Fig. 3.** Comparison of the experimental (black open circles) and configuration-averaged (blue solid lines) EXAFS  $\chi_{ca}^l(k)k^2$  for the Ti K-edge in SrTiO<sub>3</sub> and the Cu K-edge in fcc Cu.

Examples of the **feff.dat** files for SrTiO<sub>3</sub> and Cu:

```
TITLE cubic SrTiO3 a=3.905 Å
EDGE K
S02 1.0
* pot xsph fms paths genfmt ff2chi
CONTROL 0 0 0 1 1 1
PRINT 1 0 0 0 0 0
* r_scf [ l_scf n_scf ca ]
SCF 4.0 0 30 0.1
* ixc [ Vr Vi ]
EXCHANGE 0 0 0
EXAFS 20.0
RPATH 6.5
CRITERIA 2.0 2.5
POTENTIALS
* ipot z [ label l_scmf l_fms stoichiometry ]
0 22 Ti -1 -1 0
1 8 O -1 -1 3
2 38 Sr -1 -1 1
3 22 Ti -1 -1 1
ATOMS
```

```
TITLE Cu metal fcc a=3.61 Å
EDGE K
S02 1.0
* pot xsph fms paths genfmt ff2chi
CONTROL 0 0 0 1 1 1
PRINT 1 0 0 0 0 0
* r_scf [ l_scf n_scf ca ]
SCF 4.0 0 30 0.1
* ixc [ Vr Vi ]
EXCHANGE 0 0 0
EXAFS 20.0
RPATH 6.5
CRITERIA 0.0 1.5
POTENTIALS
* ipot z [ label l_scmf l_fms stoichiometry ]
0 29 Cu -1 -1 0
1 29 Cu -1 -1 1
ATOMS
```

Examples of the **pot.dat** files for SrTiO<sub>3</sub> and Cu:

# potential	potential	Atom
# in XYZ	in feff.inp (0=Ti*, 1=O, 2=Sr 3=Ti)	number
0	0	22
2	2	38
1	3	22
3	1	8

# potential	potential	Atom
# in XYZ	in feff.inp (0=Cu*, 1=Cu)	number
0	0	29
1	1	29

N.B. Lines started with # are comments.

Example of the **fdmnes.dat** file:

```
! Example of fdmnes.dat file for the K-edge absorption
```

```
Range
```

```
-20.0 1.0 -10.0 0.2 0.0 0.2 15.0 0.5 20.0 1.0 40.0 2.0 100.0
```

```
Green
```

```
Edge
```

```
K
```

```
Radius
```

```
8.0
```

```
Molecule
```

```
1.0 1.0 1.0 90. 90. 90. = a, b, c, alpha, beta, gamma
```

Example of **small XYZ** file for SrTiO<sub>3</sub>:

```
# Cluster: 1. SCF Done      -9251.24551535
2 -5.6638366219999998E+000 -1.9345635749999994E+000 -1.8965811260000001E+000
2 -5.8161227160000006E+000 -1.9902422660000001E+000 2.0225423609999993E+000
2 -5.8223570030000005E+000 1.7646858830000003E+000 -1.9921332800000000E+000
...
2 5.9074305330000003E+000 2.0739280749999995E+000 1.9143412239999993E+000
1 -7.7015225640000002E+000 9.83705000000006800E-003 -7.4649180000001536E-003
1 -3.8341469420000003E+000 -3.8993493439999996E+000 -3.9374565160000001E+000
1 -3.8725798060000005E+000 -3.9208021269999995E+000 -3.0967081000000007E-002
...
1 6.5886389000000101E-002 2.72920390000000656E-002 -3.9367556619999999E+000
0 0.0000000000000000E+000 0.0000000000000000E+000 0.0000000000000000E+000
1 -4.03979500000006233E-003 1.03095528000000077E-001 3.7966132160000008E+000
...
3 -5.7877681580000004E+000 -3.8631761129999997E+000 1.28714370000000263E-002
3 -5.8439203129999999E+000 2.2320960000001833E-003 -3.9290554370000002E+000
3 -5.8210508980000002E+000 -4.3372319999999576E-002 -3.87125260000000302E-002
...
```

Example of **small XYZ** file for Cu:

```
# Cluster: 11. Atoms. Timestep: 10
1 -1.722070000000000E+0000 -1.972020000000000E+0000 -7.313280000000000E+0000
1 1.824200000000000E+0000 -1.985650000000000E+0000 -7.259880000000000E+0000
1 -1.17989999999998E-0001 -1.282800000000000E-0001 -7.220410000000000E+0000
...
1 -2.95799999999993E-0002 1.773080000000000E+0000 -1.854810000000000E+0000
0 0.000000000000000E+0000 0.000000000000000E+0000 0.000000000000000E+0000
1 -1.874840000000000E+0000 1.791320000000000E+0000 -4.600999999999990E-0002
...
```

## Statistical analysis of EDACA results for EXAFS

After running **edaca.exe**, a set of EXAFS spectra  $\chi_i^l(k)$  ( $i = 1, \dots, n$ ) corresponding to  $n$  atomic configurations from the XYZ file is produced. They are named xt.001, xt.002, .... These files can be analysed by the **stdev.exe** code to get an idea of the convergence of the configuration-averaged EXAFS  $\chi_{ca}^l(k)$ . Four StDev() functions (Fig. 4) described below are automatically calculated for further analysis.

The configuration-averaged EXAFS signal over  $n$  MD configurations:

$$\chi_{ca}^l(k) \equiv \chi_{ca}^l(n, k) = \frac{1}{n} \sum_{i=1}^n \chi_i^l(k)$$

The configuration-averaged EXAFS signal over  $m$  MD configurations:

$$\chi_{ca}^l(m, k) = \frac{1}{m} \sum_{i=1}^m \chi_i^l(k)$$

The standard deviation (file **stdevm.txt**) of  $\chi_{ca}^l(m, k)$  from  $\chi_{ca}^l(m-1, k)$ :

$$\text{StDev}(m) = \sqrt{\frac{1}{nk} \sum_{k=1}^{nk} [\chi_{ca}^l(m, k) - \chi_{ca}^l(m-1, k)]^2}$$

The standard deviation (file **stdevav.txt**) of  $\chi_{ca}^l(m, k)$  from  $\chi_{ca}^l(n, k)$ :

$$\text{StDev}(m, n) = \sqrt{\frac{1}{nk} \sum_{k=1}^{nk} [\chi_{ca}^l(m, k) - \chi_{ca}^l(n, k)]^2}$$

The standard deviation (file **stdevx.txt**) of **two consecutive** EXAFS spectra:

$$\text{StDev}(j) = \sqrt{\frac{1}{nk} \sum_{k=1}^{nk} [\chi_j^l(k) - \chi_{j-1}^l(k)]^2}$$

The standard deviation (file **stdevxav.txt**) from the **mean** EXAFS spectrum:

$$\text{StDev}(j, n) = \sqrt{\frac{1}{nk} \sum_{k=1}^{nk} [\chi_j^l(k) - \chi_{ca}^l(n, k)]^2}$$

The **stdevav.txt** and **stdevm.txt** files are most interesting since they show the convergence process.

**N.B. the stdev.exe code cannot be used for XANES calculations!**

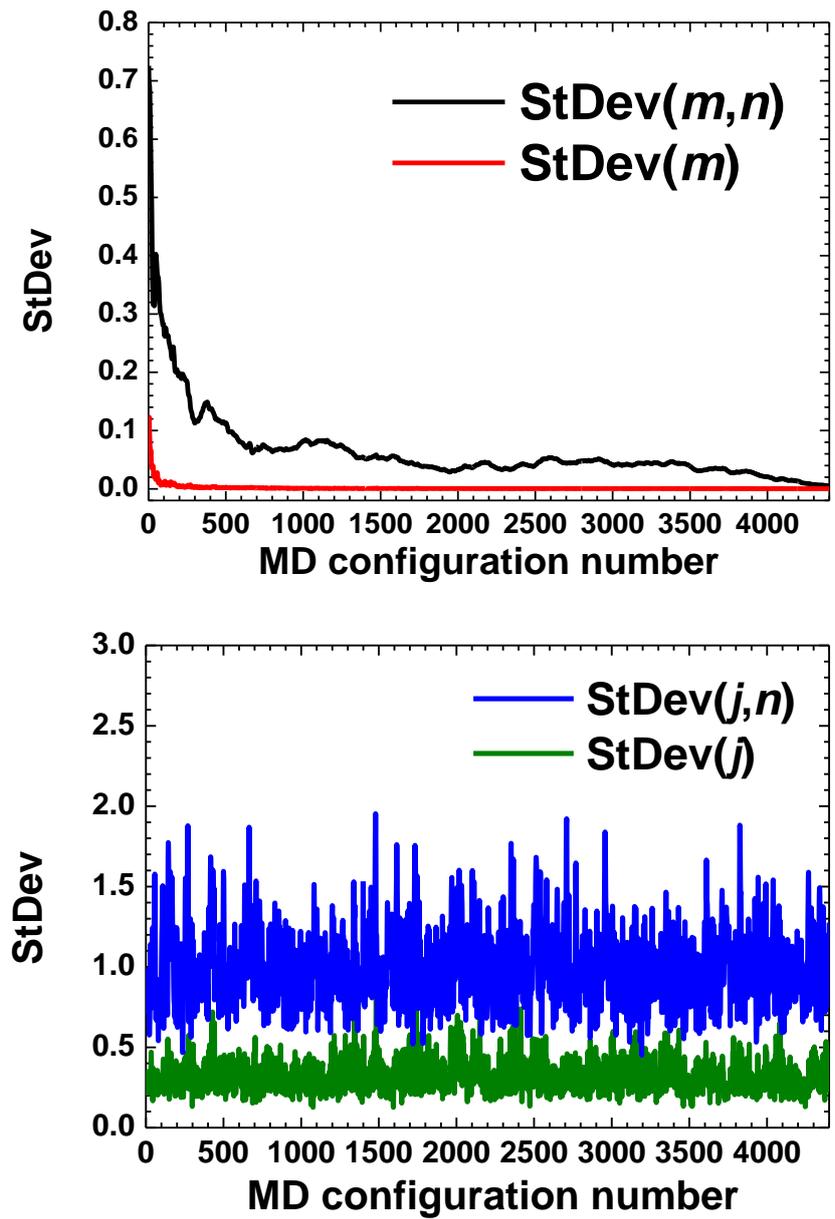


Fig. 4. Results of statistical analysis.

## Summary

Molecular Dynamics is a natural way to include disorder (static and dynamic) into EXAFS or XANES simulations taking into account multiple-scattering effects.

The MD-EXAFS approach allows one to interpret EXAFS spectra based on the multiple-scattering formalism far beyond the first coordination shell.

No structural fitting parameters are used in both MD-EXAFS and MD-XANES methods. The structural model is uniquely defined from the results of the MD simulation.

EXAFS/XANES spectra can be used as an additional physical property for interatomic potential/theory validation.

The accuracy of configuration averaged EXAFS/XANES calculations (MD-EXAFS/MD-XANES approach) is limited by the accuracy of molecular dynamics simulations (interatomic potentials).

Good choice of equilibration time, proper averaging (number of configurations (>100-1000) & time step) and simulation box size in MD simulations are crucial.

Care should be taken regarding the temperature. At low temperatures, special treatment is required, e.g. based on path-integral methodology.

EXAFS/XANES calculations for each atomic configuration are based on the FEFF or FDMNES codes, therefore all possible problems related to them should be taken into account.

## References

- [1] A. Kuzmin, A. Anspoks, A. Kalinko, J. Timoshenko, *Z. Phys. Chem.* **230** (2016) 537.
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## Appendix 1. List of EDACA applications to different materials

Materials calculated using the ab initio MD method are in **bold**, whereas those calculated using the Machine-Learning potentials are in *italics*.

Material	Reference
SrTiO <sub>3</sub>	A. Kuzmin, R.A. Evarestov, Quantum mechanics-molecular dynamics approach to the interpretation of X-ray absorption spectra, <i>J. Phys.: Condens. Matter</i> <b>21</b> (2009) 055401 (6 pp).
SrTiO <sub>3</sub>	A. Kuzmin, R.A. Evarestov, Quantum mechanics-classical molecular dynamics approach to EXAFS, <i>J. Phys.: Conf. Ser.</i> <b>190</b> (2009) 012024 (6pp).
ReO <sub>3</sub>	A. Kalinko, R.A. Evarestov, A. Kuzmin, J. Purans, Interpretation of EXAFS in ReO <sub>3</sub> using molecular dynamics simulations, <i>J. Phys.: Conf. Ser.</i> <b>190</b> (2009) 012080 (4pp).
NiO	A. Anspoks, A. Kuzmin, A. Kalinko, J. Timoshenko, Probing NiO nanocrystals by EXAFS spectroscopy, <i>Solid State Commun.</i> <b>150</b> (2010) 2270-2274.
Ge	J. Timoshenko, A. Kuzmin, J. Purans, Molecular dynamics simulations of EXAFS in germanium, <i>Centr. Eur. J. Phys.</i> <b>9</b> (2011) 710-715.
LaCoO <sub>3</sub>	A. Kuzmin, V. Efimov, E. Efimova, V. Sikolenko, S. Pascarelli, I. O. Troyanchuk, Interpretation of the Co K-edge EXAFS in LaCoO <sub>3</sub> using molecular dynamics simulations, <i>Solid State Ionics</i> <b>188</b> (2011) 21-24.
NiO	A. Anspoks, A. Kuzmin, Interpretation of the Ni K-edge EXAFS in nanocrystalline nickel oxide using molecular dynamics simulations, <i>J. Non-Cryst. Solids</i> <b>357</b> (2011) 2604-2610.
NiO	A. Anspoks, A. Kalinko, R. Kalendarev, A. Kuzmin, Atomic structure relaxation in nanocrystalline NiO studied by EXAFS spectroscopy: Role of nickel vacancies, <i>Phys. Rev. B</i> <b>86</b> (2012) 174114:1-11.
NiO	A. Anspoks, A. Kalinko, R. Kalendarev, A. Kuzmin, Probing vacancies in NiO nanoparticles by EXAFS and molecular dynamics simulations, <i>J. Phys.: Conf. Ser.</i> <b>430</b> (2013) 012027:1-4.
CaWO <sub>4</sub> SrWO <sub>4</sub> BaWO <sub>4</sub>	A. Kalinko and A. Kuzmin, Interpretation of EXAFS in scheelite-type AWO <sub>4</sub> (A= Ca, Sr, Ba) compounds using molecular dynamics simulations, <i>J. Phys.: Conf. Ser.</i> <b>430</b> (2013) 012075:1-4.
NiO	A. Anspoks, A. Kalinko, R. Kalendarev, A. Kuzmin, Local structure relaxation in nanocrystalline Ni <sub>1-x</sub> O thin films, <i>Thin Solid Films</i> <b>553</b> (2014) 58-62.
ZnO	J. Timoshenko, A. Anspoks, A. Kalinko, A. Kuzmin, Temperature dependence of the local structure and lattice dynamics of wurtzite-type ZnO, <i>Acta Mater.</i> <b>79</b> (2014) 194-202.
ZnO	J. Timoshenko, A. Anspoks, A. Kalinko, A. Kuzmin, Local structure and dynamics of wurtzite-type ZnO from simulation-based EXAFS analysis, <i>Phys. Status Solidi (c)</i> <b>11</b> (2014) 1472-1475.
Y <sub>2</sub> O <sub>3</sub>	K. Lazdins, A. Kuzmin, Local structure and lattice dynamics of cubic Y <sub>2</sub> O <sub>3</sub> : an x-ray absorption spectroscopy study, <i>IOP Conf. Ser.: Mater. Sci. Eng.</i> <b>77</b> (2015) 012031:1-5.

SrTiO <sub>3</sub> ZnO	A. Kuzmin, A. Anspoks, A. Kalinko, J. Timoshenko, The use of x-ray absorption spectra for validation of classical force-field models, <i>Z. Phys. Chem.</i> <b>230</b> (2016) 537-549.
ScF <sub>3</sub>	D. Bocharov, M. Krack, A. Kalinko, J. Purans, F. Rocca, S. E. Ali, A. Kuzmin, Ab initio molecular dynamics simulations of the Sc K-edge EXAFS of scandium trifluoride, <i>J. Phys.: Conf. Ser.</i> <b>712</b> (2016) 012009:1-4.
UO <sub>2</sub>	D. Bocharov, M. Chollet, M. Krack, J. Bertsch, D. Grolimund, M. Martin, A. Kuzmin, J. Purans, E. Kotomin, Interpretation of the U L <sub>3</sub> -edge EXAFS in uranium dioxide using molecular dynamics and density functional theory simulations, <i>J. Phys.: Conf. Ser.</i> <b>712</b> (2016) 012091:1-4.
Y <sub>2</sub> O <sub>3</sub>	I. Jonane, K. Lazdins, J. Timoshenko, A. Kuzmin, J. Purans, P. Vladimirov, T. Gräning, J. Hoffmann, Temperature-dependent EXAFS study of the local structure and lattice dynamics in cubic Y <sub>2</sub> O <sub>3</sub> , <i>J. Synchrotron Rad.</i> <b>23</b> (2016) 510-518.
Cu <sub>3</sub> N	J. Timoshenko, A. Anspoks, A. Kalinko, A. Kuzmin, Local structure of copper nitride revealed by EXAFS spectroscopy and reverse Monte Carlo/evolutionary algorithm approach, <i>Phys. Scr.</i> <b>91</b> (2016) 054003 (11pp).
FeF <sub>3</sub>	I. Jonane, J. Timoshenko, A. Kuzmin, Atomistic simulations of the Fe K-edge EXAFS in FeF <sub>3</sub> using molecular dynamics and reverse Monte Carlo methods, <i>Phys. Scr.</i> <b>91</b> (2016) 104001 (6pp).
CaWO <sub>4</sub> SrWO <sub>4</sub> BaWO <sub>4</sub>	A. Kalinko, A. Bauer, J. Timoshenko, A. Kuzmin, Molecular dynamics and reverse Monte Carlo modeling of scheelite-type AWO <sub>4</sub> (A=Ca, Sr, Ba) W L <sub>3</sub> -edge EXAFS spectra, <i>Phys. Scr.</i> <b>91</b> (2016) 114001 (9pp).
UO <sub>2</sub>	D. Bocharov, M. Chollet, M. Krack, J. Bertsch, D. Grolimund, M. Martin, A. Kuzmin, J. Purans, E. Kotomin, Analysis of the U L <sub>3</sub> -edge X-ray absorption spectra in UO <sub>2</sub> using molecular dynamics simulations, <i>Prog. Nucl. Energy</i> <b>94</b> (2017) 187-193.
W	I. Jonane, A. Anspoks, A. Kuzmin, Advanced approach to the local structure reconstruction and theory validation on the example of the W L <sub>3</sub> -edge extended X-ray absorption fine structure of tungsten, <i>Modelling Simul. Mater. Sci. Eng.</i> <b>26</b> (2018) 025004 (11 pp).
Cu <sub>3</sub> N	D. Bocharov, A. Anspoks, J. Timoshenko, A. Kalinko, M. Krack, A. Kuzmin, Interpretation of the Cu K-edge EXAFS spectra of Cu <sub>3</sub> N using ab initio molecular dynamics, <i>Rad. Phys. Chem.</i> <b>175</b> (2020) 108100.
ScF <sub>3</sub>	D. Bocharov, M. Krack, Yu. Rafalskij, A. Kuzmin, J. Purans, Ab initio molecular dynamics simulations of negative thermal expansion in ScF <sub>3</sub> : the effect of the supercell size, <i>Comp. Mater. Sci.</i> <b>171</b> (2020) 109198.
ZnO	D. Bocharov, I. Pudza, K. Klementiev, M. Krack, A. Kuzmin, Study of high-temperature behaviour of ZnO by ab initio molecular dynamics simulations and X-ray absorption spectroscopy, <i>Materials</i> <b>14</b> (2021) 5206.
W Mo Cu Ni	A. V. Shapeev, D. Bocharov, A. Kuzmin, Validation of moment tensor potentials for fcc and bcc metals using EXAFS spectra, <i>Comput. Mater. Sci.</i> <b>210</b> (2022) 111028.

<b>MoS<sub>2</sub></b>	I. Pudza, D. Bocharov, A. Anspoks, M. Krack, A. Kalinko, E. Welter, A. Kuzmin, Unraveling the interlayer and intralayer coupling in two-dimensional layered MoS <sub>2</sub> by X-ray absorption spectroscopy and ab initio molecular dynamics simulations, Mater. Today Commun. <b>35</b> (2023) 106359.
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