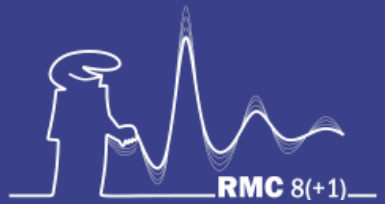

PROBING NIO LATTICE DYNAMICS BY X-RAY ABSORPTION SPECTROSCOPY COMBINED WITH REVERSE MONTE CARLO METHOD

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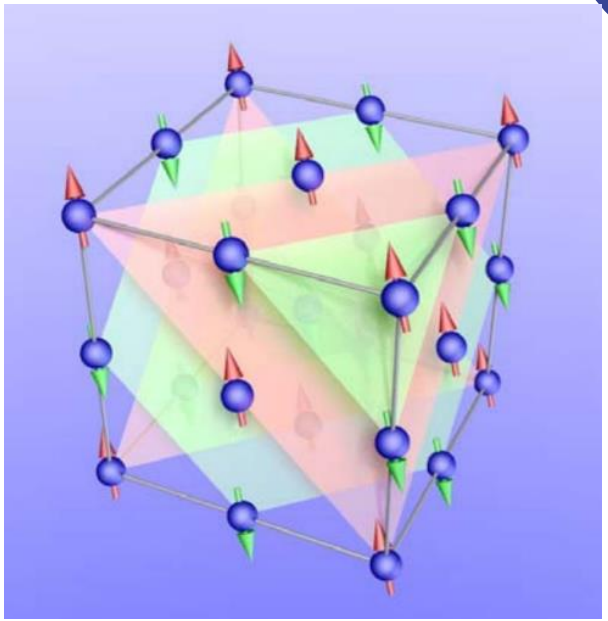
Aleksandr Kalinko, Alexei Kuzmin

MOTIVATION

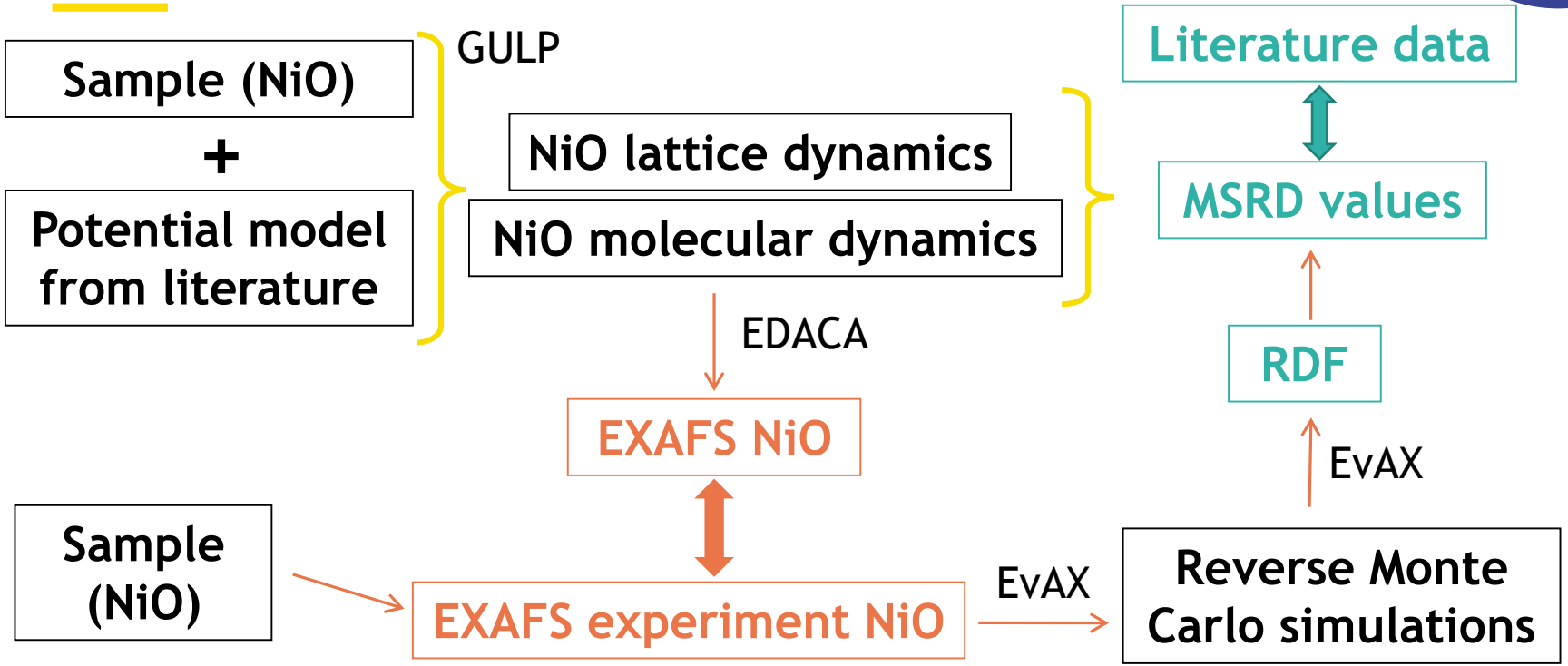
The lattice dynamics of the Mott-Hubbard insulator NiO with a magnetic Néel phase transition at 523 K has been the subject of numerous studies for many years.
Until now, the accent of most investigations has been paid to phonon frequencies.

Does lattice distortion influence atomic interactions?

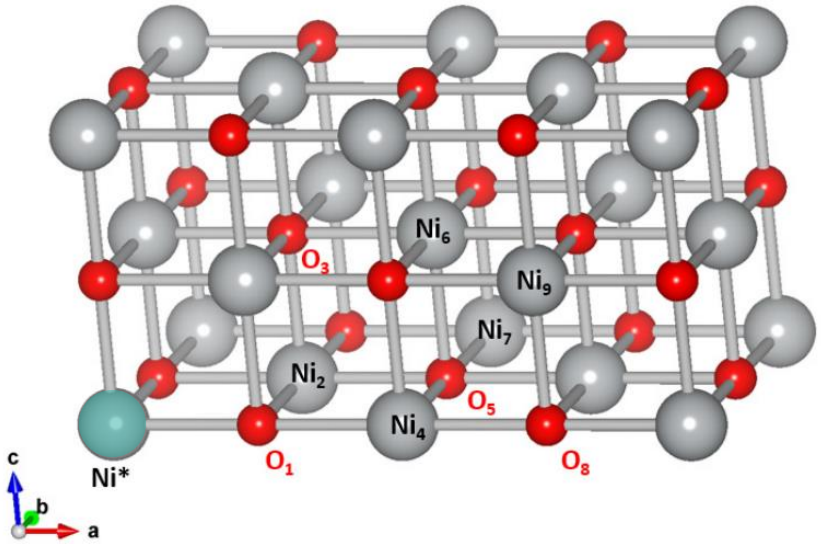


Antiferromagnetic structure of NiO with ferromagnetic planes, O atoms are not shown. Blue spheres - Ni atoms, arrows – spins

OUTLOOK

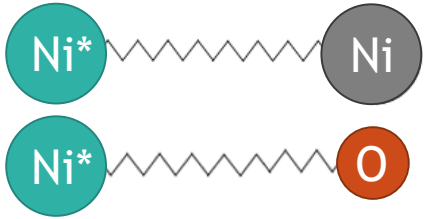

Sample (NiO)
+
Potential model from literature
GULP
NiO lattice dynamics
NiO molecular dynamics
Literature data
MSRD values
RDF
EvAX
EDACA
EXAFS NiO
Sample (NiO)
EXAFS experiment NiO
EvAX
Reverse Monte Carlo simulations

NiO STRUCTURE AND LATTICE DYNAMICS MODEL

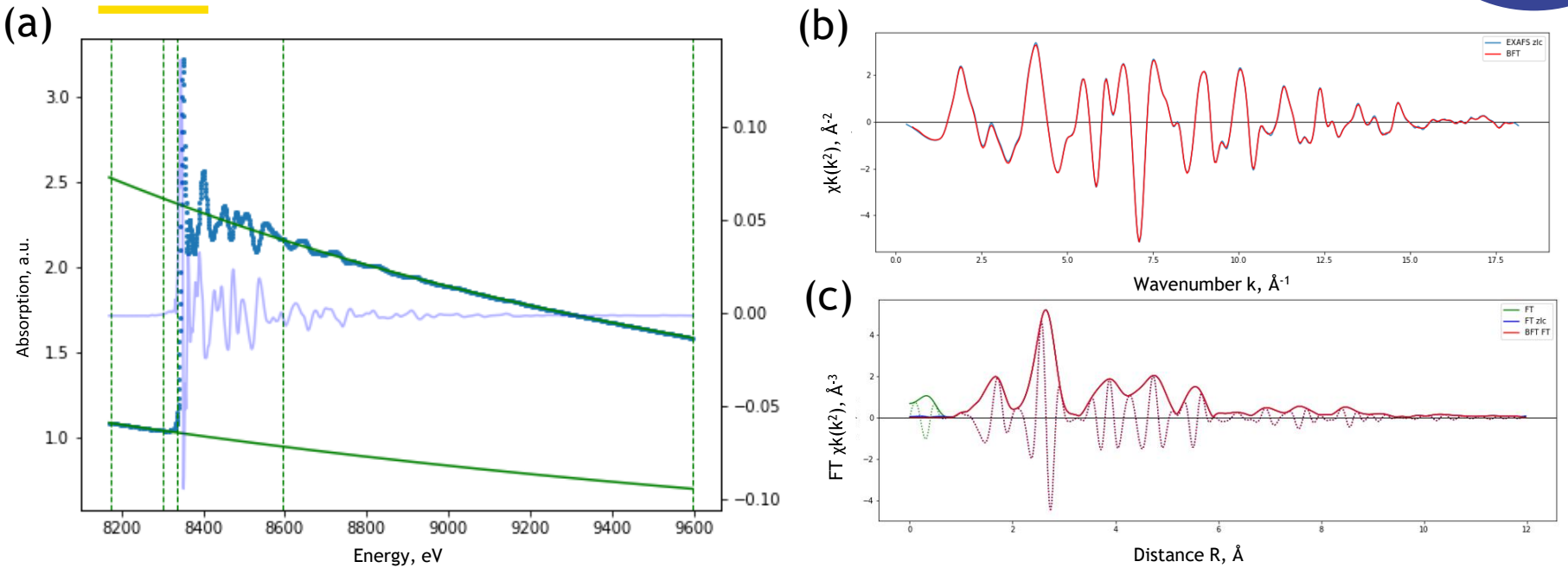


MSRD(NiO) = MSD (Ni) + MSD (O) – DCF(NiO)

DCF – correlation function



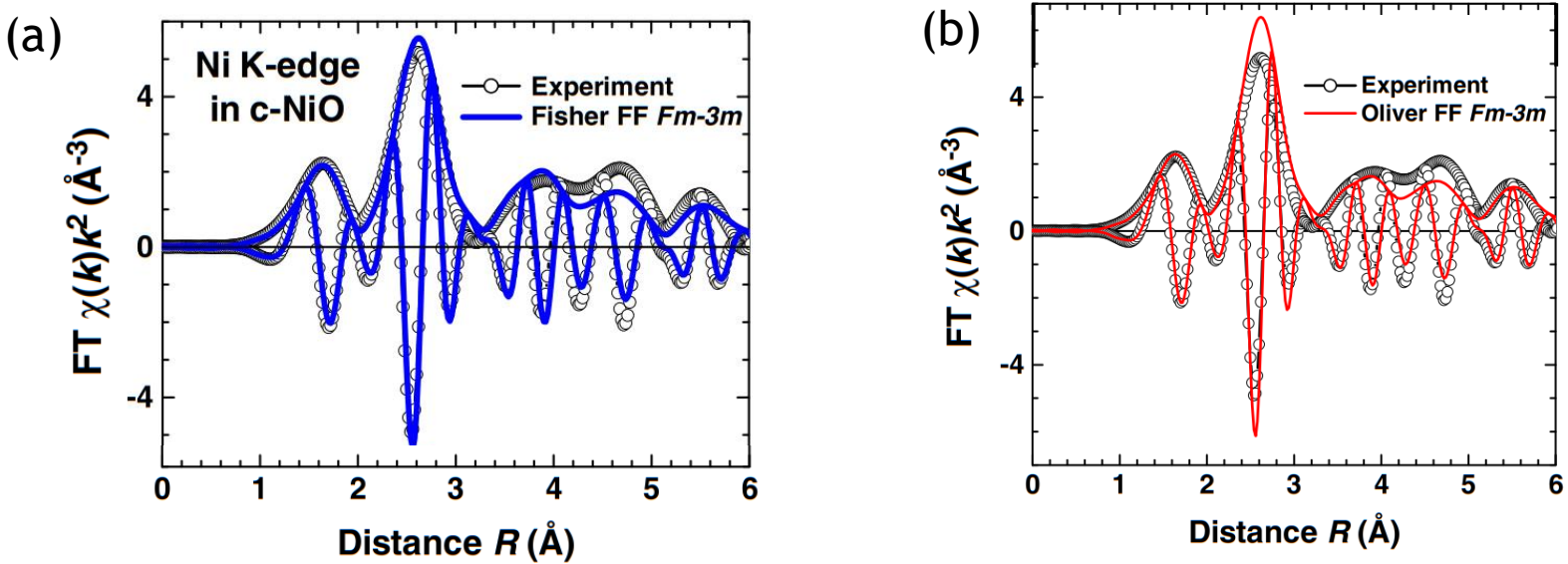
X-RAY ABSORPTION EXPERIMENT



Ni K-edge XAS of NiO at 300K (a), Ni K-edge EXAFS $\chi(k)k^2$ spectrum (b), FT of Ni K-edge EXAFS spectrum (c).

A. Kalinko, XAESA v0.05. https://github.com/aklnk/xaesa/

POTENTIAL MODEL FOR LATTICE DYNAMICS



Comparison of the experimental (open circles) and MD calculated (solid line) Fourier transforms of Ni K-edge EXAFS $\chi(k)k^2$ spectra of NiO. Fisher FF potential (a) and Oliver FF potential (b).

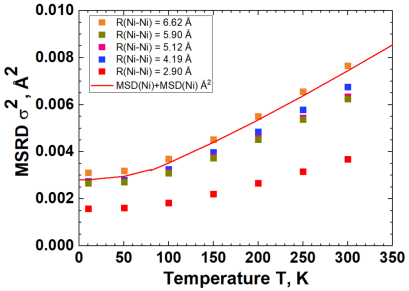
A. Anspoks, A. Kuzmin, J. Non-Cryst. Solids 357 (2011) 2604-2610

LATTICE DYNAMICS CALCULATIONS

- ✓ Fisher potential model parameters
- ✓ Cell parameters
- ✓ Shrink factor
- ✓ Temperature

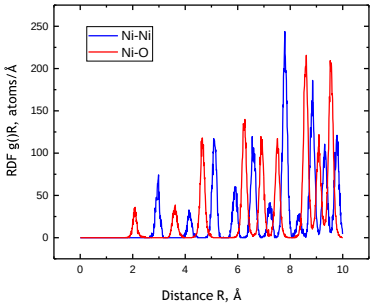
→ GULP →

MSRDs



J. D. Gale, A.L. Rohl, The General Utility Lattice Program (GULP). Molecular Simulations, 2003, 291–341.

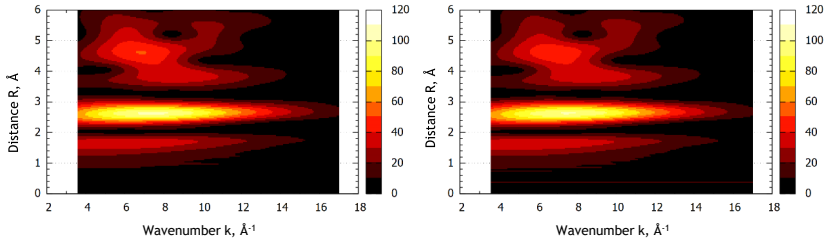
RMC SIMULATIONS OF NIO



- ✓ Experiment – Ni K-edge EXAFS spectrum
- ✓ Structure Fm-3m
- ✓ Supercell parameters (4x4x4)
- ✓ Absorbing atom - Ni

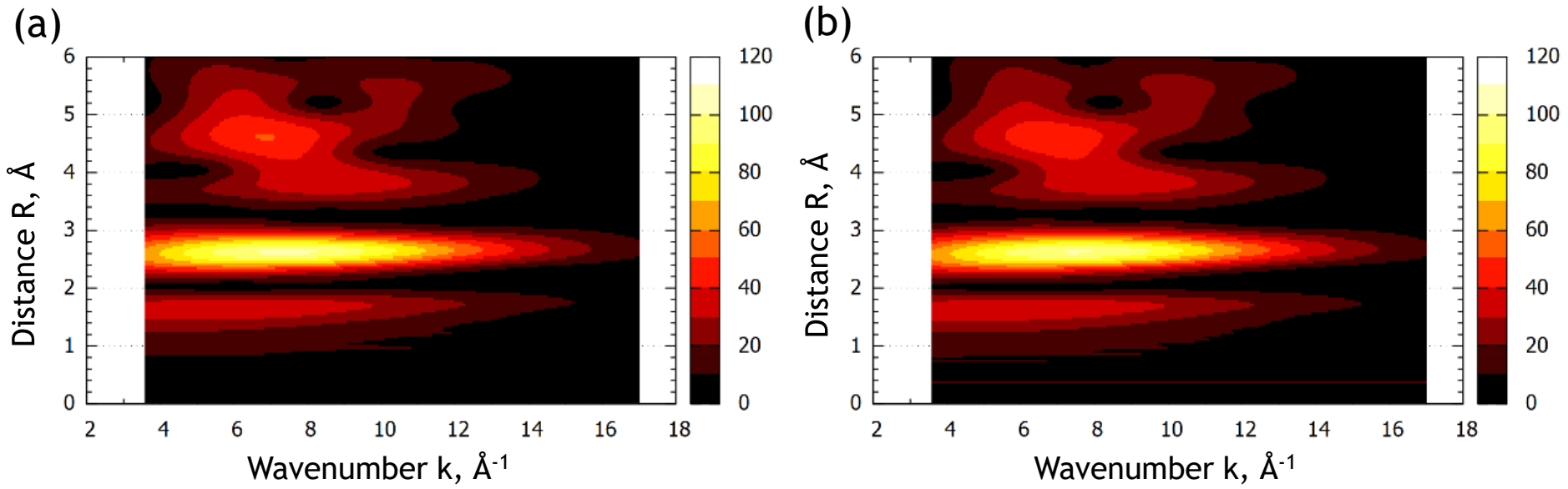
→ EvAX → RDF → MSRDs

↘ Simulated EXAFS spectrum and its comparison to experiment



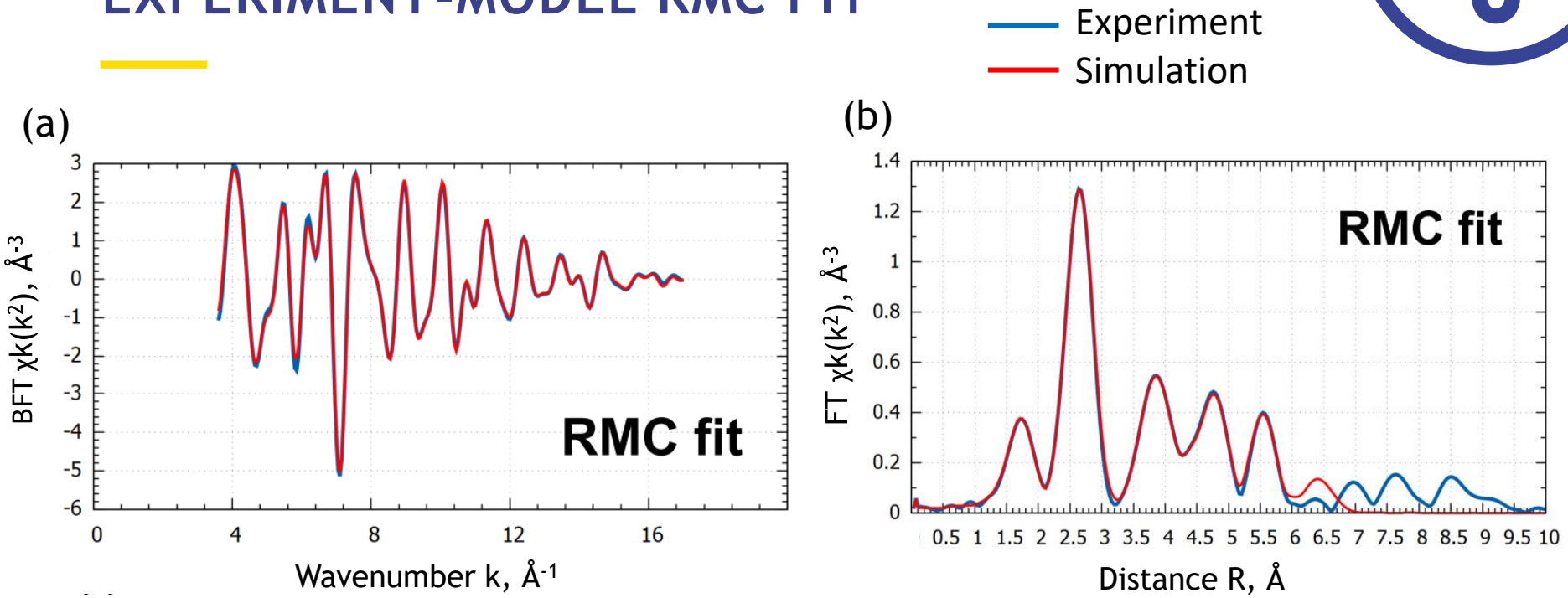
J. Timoshenko, A. Kuzmin, J. Purans, EXAFS study of hydrogen intercalation into ReO3 using the evolutionary algorithm. J. Phys.: Condens. Matter, 2014.

EXPERIMENT-MODEL RMC FIT



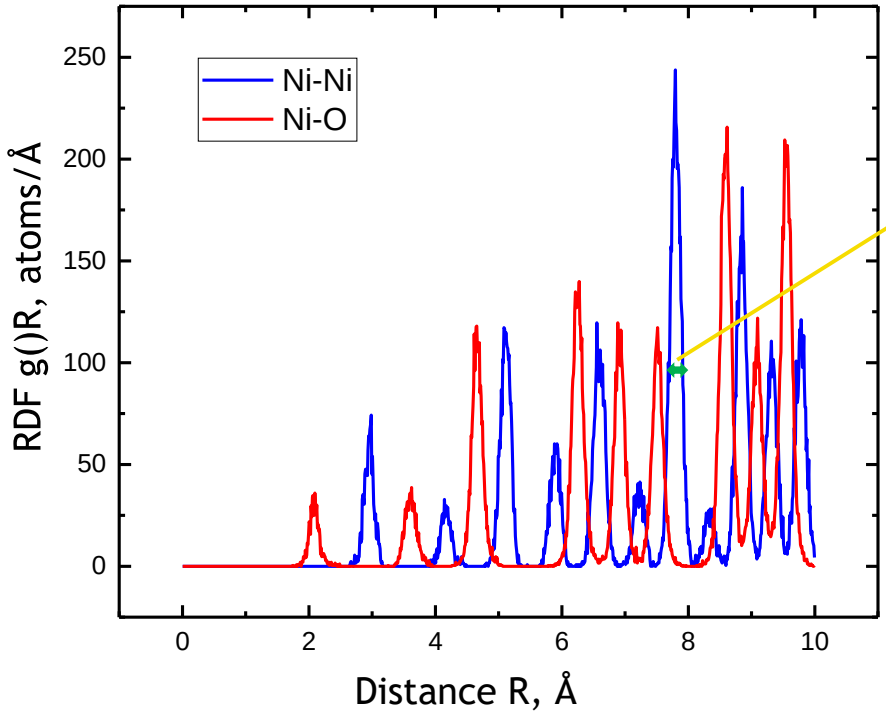
Comparison of the RMC simulated (a) and experiment (b) WT of Ni K-edge EXAFS $\chi(k)k^2$ spectra at 300 K.

EXPERIMENT-MODEL RMC FIT



Comparison of the experimental (blue line) and RMC simulated (red lines) Ni K-edge EXAFS spectra $\chi(k)k^2$ (a) and their Fourier transforms (b) at 300 K.

RADIAL DISTRIBUTION FUNCTIONS FROM RMC

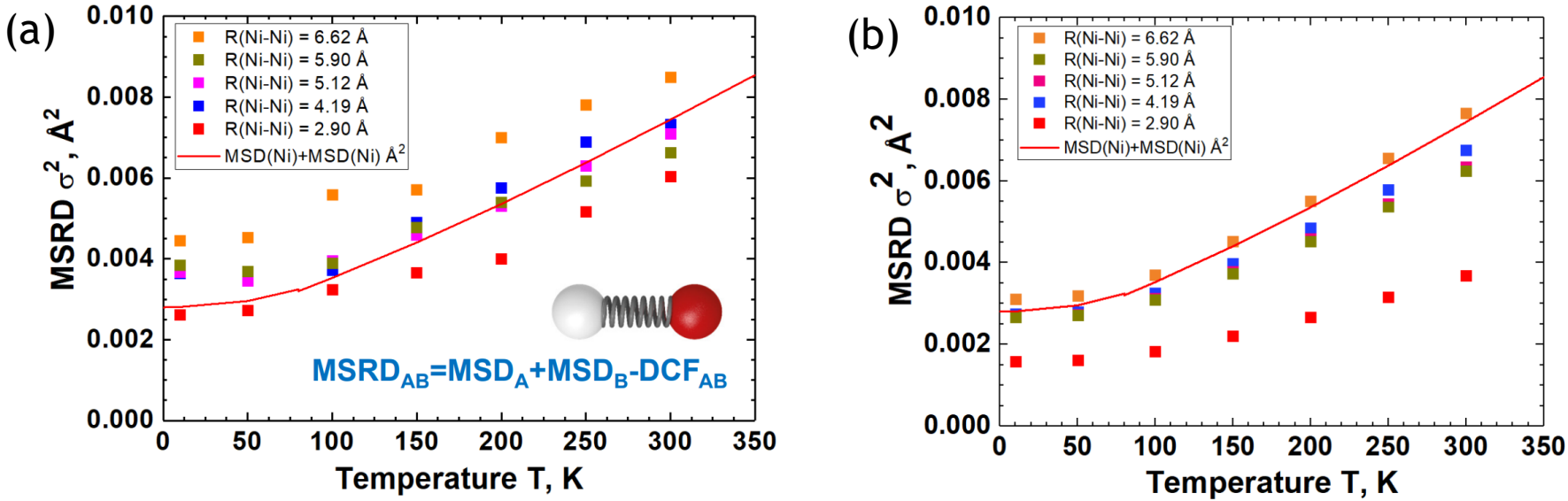


w – FWHM

MSRD

RDF as a result of RMC simulation for Ni K-edge EXAFS $\chi(k)k^2$ spectra at 300 K.

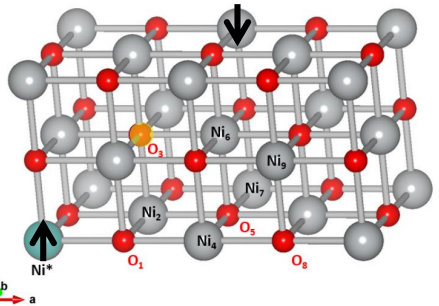
MSRD VALUES FOR Ni-Ni PAIRS

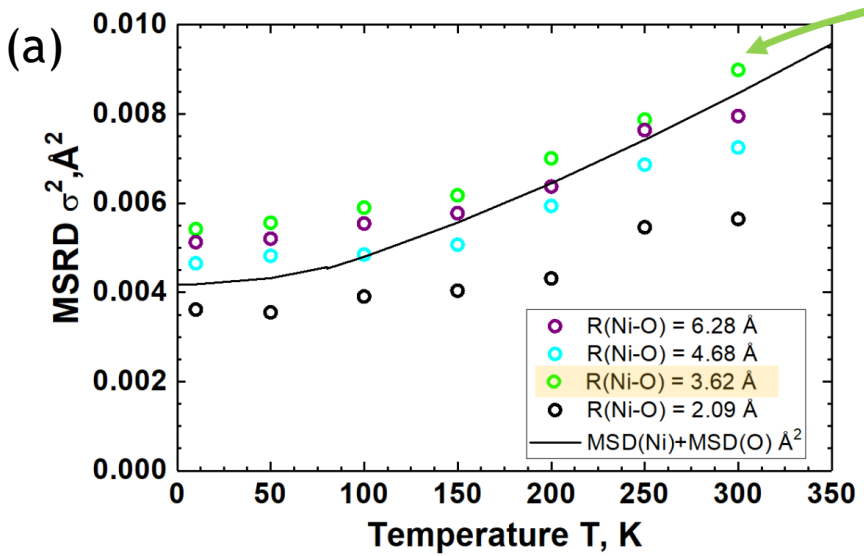


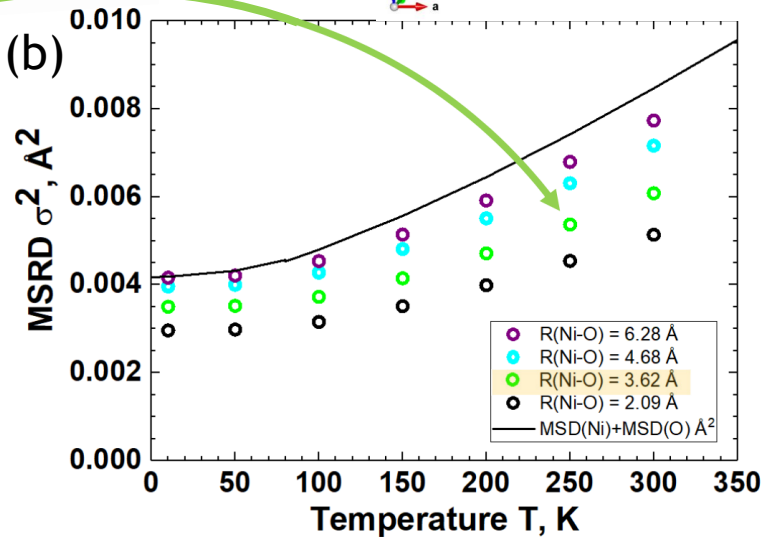
MSRD values of Ni-Ni pairs as a function of temperature. Results of the RMC simulations (a) and lattice dynamics calculations (b).

H. X. Gao, L. M. Penga, J. M. Zuo, Acta Cryst. A 55 (1999) 1014-1025

MSRD VALUES FOR Ni-O PAIRS







MSRD values of Ni-O pairs as a function of temperature. Results of the RMC simulations (a) and lattice dynamics calculations (b).

H. X. Gao, L. M. Penga, J. M. Zuo, Acta Cryst. A 55 (1999) 1014-1025

CONCLUSIONS

- Reverse Monte Carlo simulations describe the Ni K-edge EXAFS spectra of NiO better than molecular dynamics calculations.
- For large interatomic distances (>5-6 Å), the correlation in atomic motion for the Ni-Ni and Ni-O pairs is reduced, as expected.
- Nevertheless, the correlation of atomic motion is also significantly reduced between the Ni and O3 atoms located along the <111> crystallographic direction.

THANK YOU!

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The financial support from the Latvian Council of Science project No. lzp-2022/1-0608 is greatly acknowledged.