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Coupled Magnetic Spin Dynamics and Molecular Dynamics in a Massively Parallel Framework

PRESENTED BY

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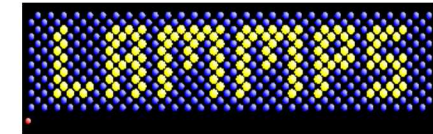
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● I Introduction

- ▶ Motivations for this development.
- ▶ Basic idea of the methodology.
- ▶ First successes, limitations.



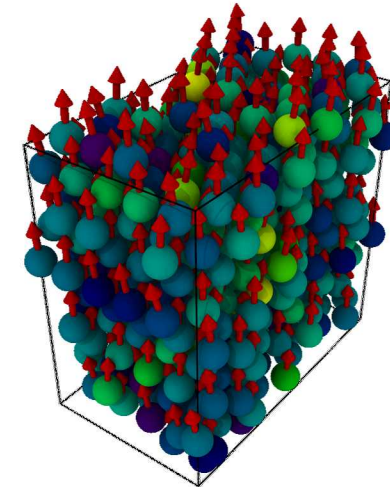
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● II Implementation

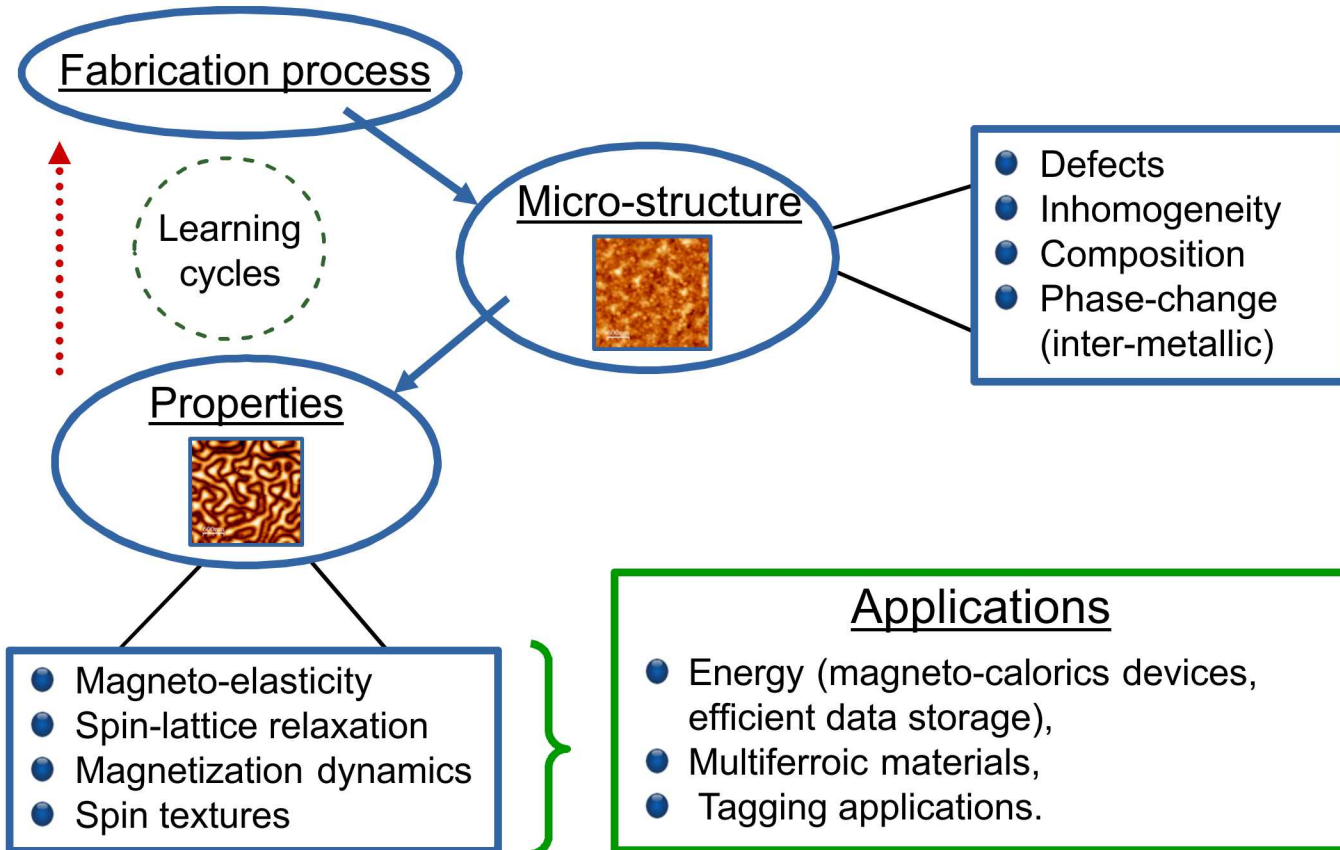
- ▶ Symplectic SD-MD serial algorithm.
- ▶ Accounting for the temperature effects.
- ▶ Spin-lattice relaxation results.
- ▶ Parallel implementation, accuracy and scalability.

● III Finally doing some physics

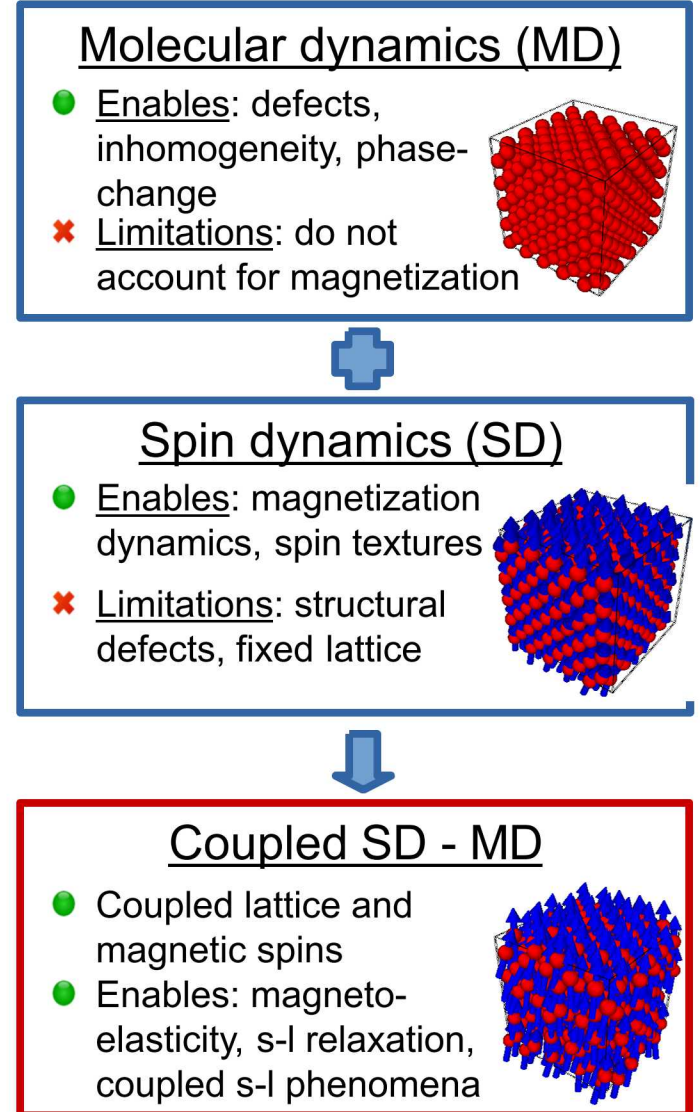
- ▶ Implemented magnetic interactions.
- ▶ Parametrization of an interaction (exchange).
- ▶ Example 1: BFO.
- ▶ Example 2: Magnetoelasticity of hcp cobalt.



A computational model coupling micro-structure and magnetic properties



Development of a model enhancing those learning cycles, and enabling the study of magnetoelastic effects and of the influence of the micro-structure on the magnetic properties.

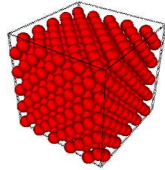


Methodology, first successes, and some current limitations

Methodology

⋮

Molecular dynamics (MD)



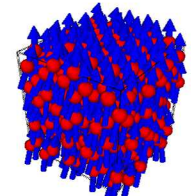
$$\left. \begin{aligned} \frac{\partial \mathbf{r}_i}{\partial t} &= \mathbf{v}_i \\ \frac{\partial \mathbf{v}_i}{\partial t} &= \mathbf{F}_i(\mathbf{r}_{ij}, \mathbf{s}_{i,j}) \end{aligned} \right\}$$

$$\mathcal{H} = \underbrace{\sum_{i=1}^N \frac{|\mathbf{p}_i|^2}{2m_i} + \sum_{i,j=1}^N V(r_{ij})}_{\text{MD Hamiltonian}}$$



Coupled SD - MD

$$\left\{ \begin{aligned} \frac{\partial \mathbf{r}_i}{\partial t} &= \mathbf{v}_i \\ \frac{\partial \mathbf{v}_i}{\partial t} &= \mathbf{F}_i(\mathbf{r}_{ij}, \mathbf{s}_{i,j}) \\ \frac{\partial \mathbf{s}_i}{\partial t} &= \boldsymbol{\omega}_i \times \mathbf{s}_i \end{aligned} \right.$$



$$\mathbf{H}_{sl} = \underbrace{\sum_{i=1}^N \frac{|\mathbf{p}_i|^2}{2m_i} + \sum_{i,j=1}^N V(r_{ij})}_{\text{MD Hamiltonian}} - \underbrace{\sum_{i,j,i \neq j}^N J(r_{ij}) \mathbf{s}_i \cdot \mathbf{s}_j - \mu_B \mu_0 \sum_{i=0}^N g_i \mathbf{s}_i \cdot \mathbf{H}_{ext}}_{\text{Spin-lattice coupling}}$$

First successes:

- Simulation of magneto-caloric effects
 - ▶ Ma, P. W., & Dudarev, S. L. (2014). Dynamic magnetocaloric effect in bcc iron and hcp gadolinium. *Physical Review B*, 90(2), 024425.
- Influence of defects on the magnetism of bcc iron
 - ▶ Mudrick, M., et al. Combined molecular and spin dynamics simulation of bcc iron with lattice vacancies. In *Journal of Physics: Conference Series* (Vol. 921, No. 1, p. 012007).
 - ▶ Wen, H., Ma, P. W., & Woo, C. H. (2013). Spin-lattice dynamics study of vacancy formation and migration in ferromagnetic BCC iron. *Journal of Nuclear Materials*, 440(1-3), 428-434.

Some current limitations:


- ✗ No release in an open-source MD code
 - preventing from the development of a large user base
- ✗ Insufficient level of parallelization
 - does not allow the simulation of large magnetic devices
- ✗ Simulation of the spin-orbit coupling
 - many technological application rely on magnetoelasticity
- ✗ Simulation of the long-range dipolar interaction
 - Unable to stabilize the domain structure of actual magnets



- Summary of the introduction:

- ▶ Necessity to develop models coupling micro-structure and magnetic properties.
- ▶ Four limitations of the former implementations were analyzed.

- Next section: Implementation of coupled SD-MD (CS part)

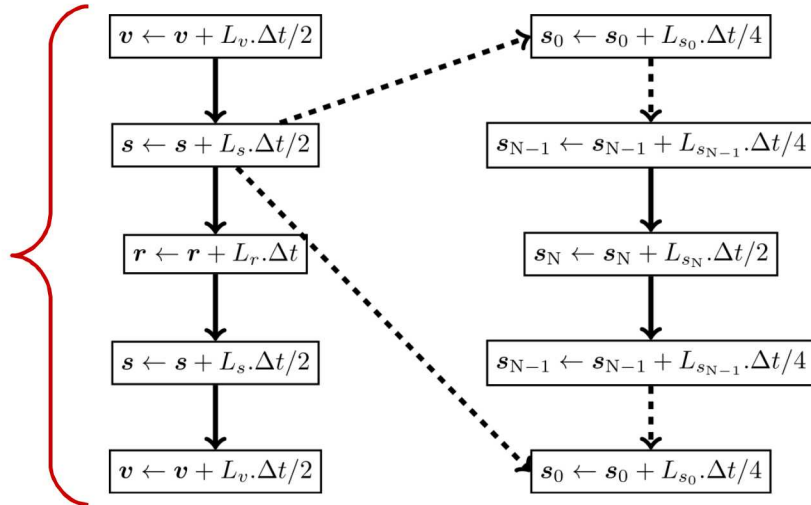
- ▶ Implementation symplectic algorithm for coupled SD-MD.
 - ▶ Parallelization, and properties (scalability and accuracy)
- 

Symplectic implementation of SD-MD

6

Symplectic spin-lattice integration scheme:

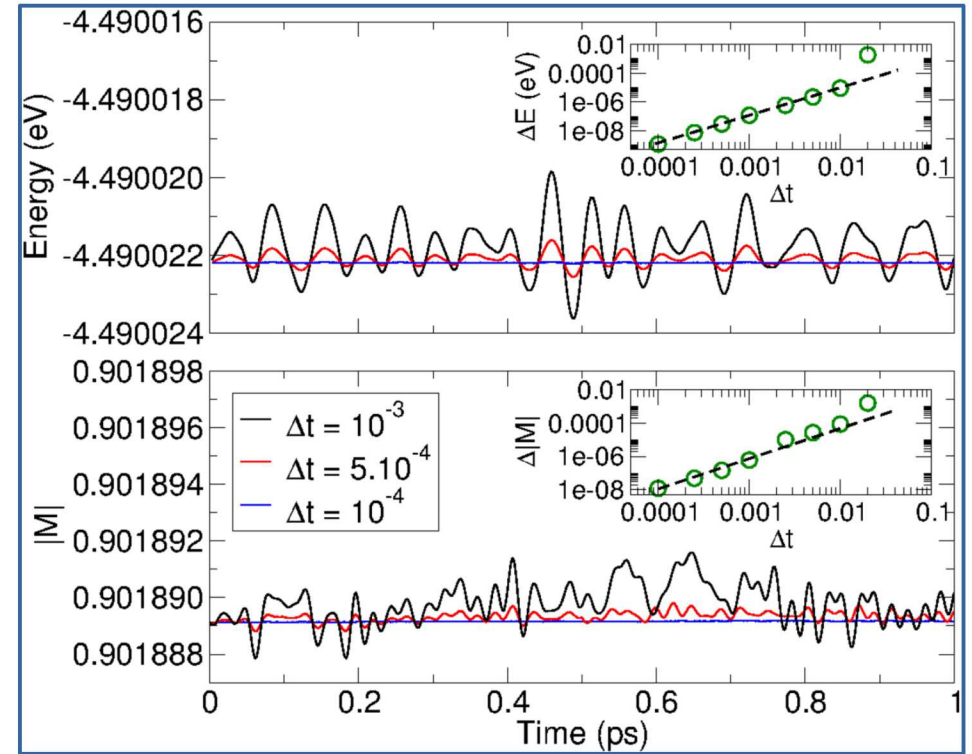
- Suzuki-Trotter decomposition applied to the global integration, and to the spin:



- With L_v , L_s and L_r being the advance operators of the velocities, spins and positions.
- Each spin advance is performed according to a geometric expression accurate in Δt .

Accuracy of the numerical scheme:

- Varying the value of the timestep Δt from 0.1 fs to 0.2 ps.



- Numerical accuracy Δt^2 (as expected).

▶ Stochastic equations for the spin-lattice system

- Two Langevin thermostats are applied to the lattice and spins systems:

$$\frac{d\mathbf{r}_i}{dt} = \frac{\mathbf{p}_i}{m_i}$$

$$\frac{d\mathbf{p}_i}{dt} = \sum_{j, i \neq j}^N \left[-\frac{dV(r_{ij})}{dr_{ij}} + \frac{dJ(r_{ij})}{dr_{ij}} \mathbf{s}_i \cdot \mathbf{s}_j \right] \mathbf{e}_{ij} - \frac{\gamma_L}{m_i} \mathbf{p}_i + \mathbf{f}(t)$$

$$\frac{d\mathbf{s}_i}{dt} = \frac{1}{(1 + \lambda^2)} \left((\boldsymbol{\omega}_i + \boldsymbol{\eta}(t)) \times \mathbf{s}_i + \lambda \mathbf{s}_i \times (\boldsymbol{\omega}_i \times \mathbf{s}_i) \right)$$

- The magnetic random torque is defined as a white-noise drawn from a Gaussian distribution:

$$\begin{aligned} \langle \boldsymbol{\eta}(t) \rangle &= \mathbf{0} \\ \langle \eta_\alpha(t) \eta_\beta(t') \rangle &= 2D_S \delta_{\alpha\beta} \delta(t - t') \\ \text{with } D_S &= \frac{2\pi\lambda k_B T}{\hbar} \end{aligned}$$

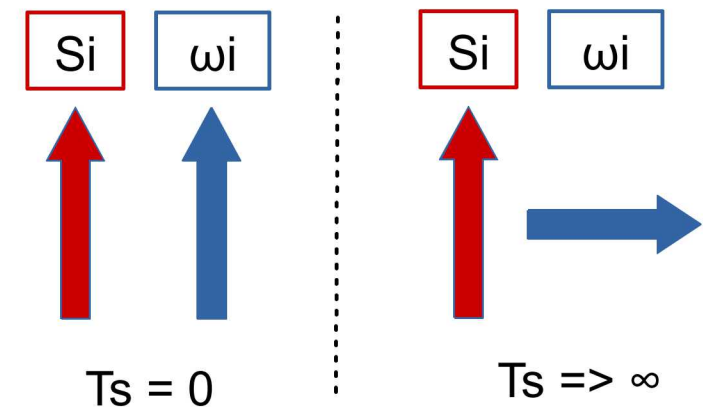
▶ Spin and lattice temperatures

- Spin and lattice temperatures are defined as follow:

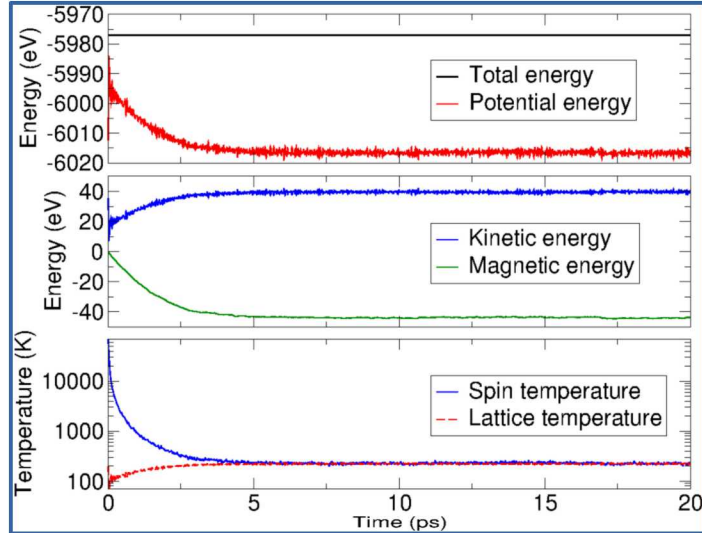
$$T_L = \frac{2}{3 N k_B} \sum_{i=1}^N \frac{|\mathbf{p}_i|^2}{2m_i}$$

$$T_S = \frac{\hbar}{2k_B} \frac{\sum_{i=1}^N |\mathbf{s}_i \times \boldsymbol{\omega}_i|^2}{\sum_{i=1}^N \mathbf{s}_i \cdot \boldsymbol{\omega}_i}$$

- This spin temperature T_S is a measure of the magnetic disorder in the system:

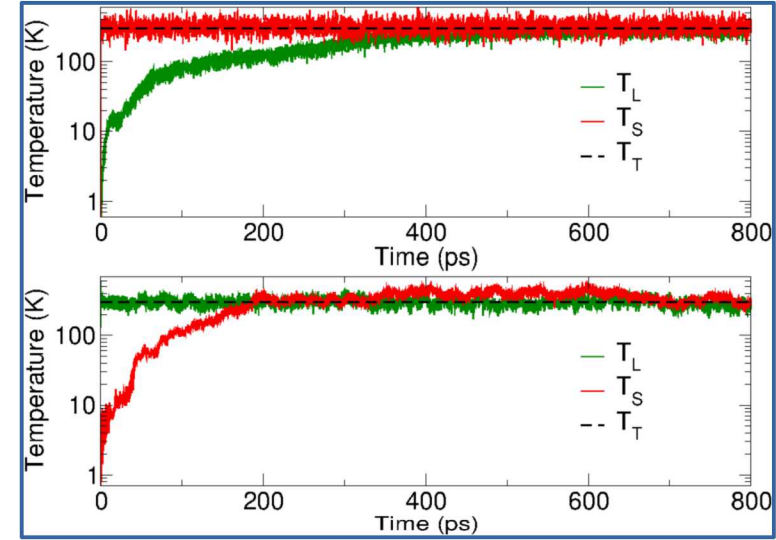


Spin-lattice relaxation, NVE calculation



- Parameters of fcc-cobalt.
- Preservation of the total energy as the kinetic, potential and magnetic energy are compensating each others.
- Relaxation and equilibration of the spin and lattice temperatures

Spin-lattice relaxation, NVT calculation



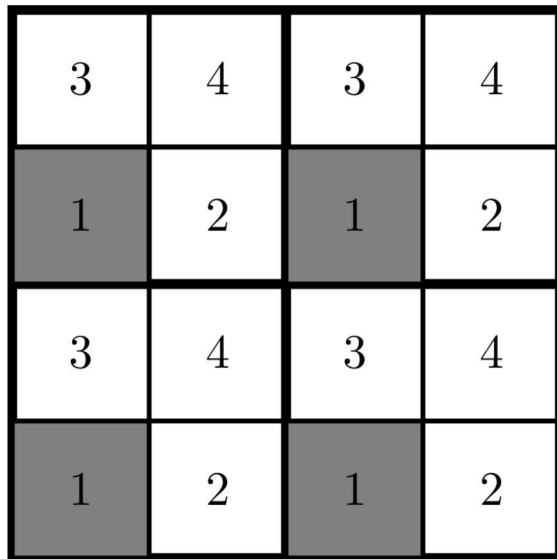
- Parameters of fcc-cobalt
- Langevin thermostat applied to the spins and to the lattice
- Relaxation and equilibration of the spin and lattice temperatures.

Sectoring algorithm for scalable SD-MD

9

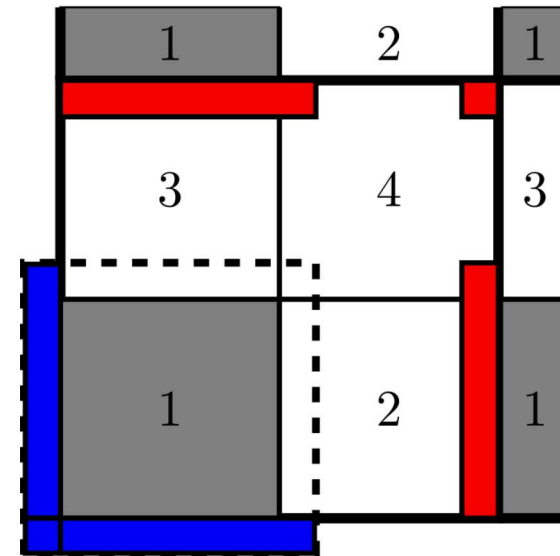
- A symplectic sectoring algorithm for massively parallel SD-MD simulations.

Sectoring algorithm for a 2D sample



- LAMMPS divide the simulation cell into domains.
- Each domain is divided into sectors (4 in 2D, 8 in 3D).
- Spins in sectors with the same label are sure not to be neighbors: they can be updated simultaneously.

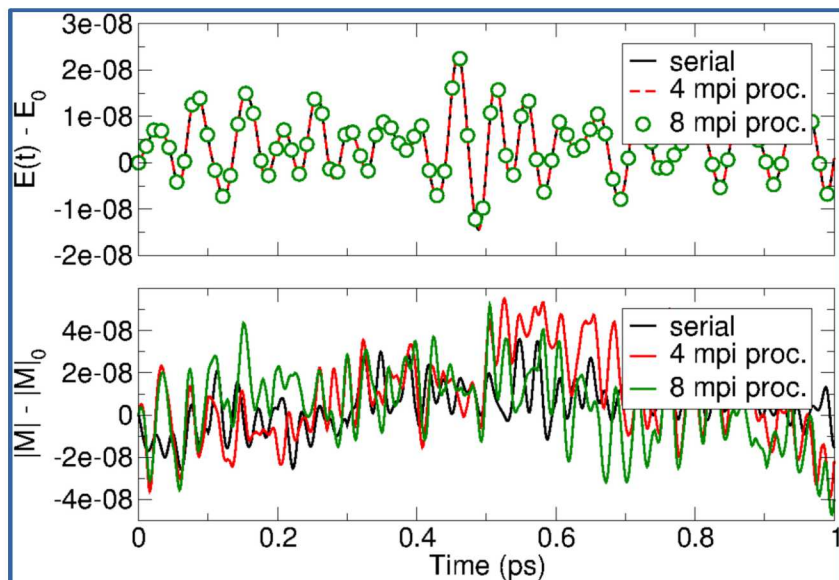
Communication operations between sectors



- Focus on the bottom left domain of the previous figure.
- Communication operation after the advance operation of the spins in a sectors labeled 1.
- Red areas have to be sent, the blue ones have to be received from other domains.

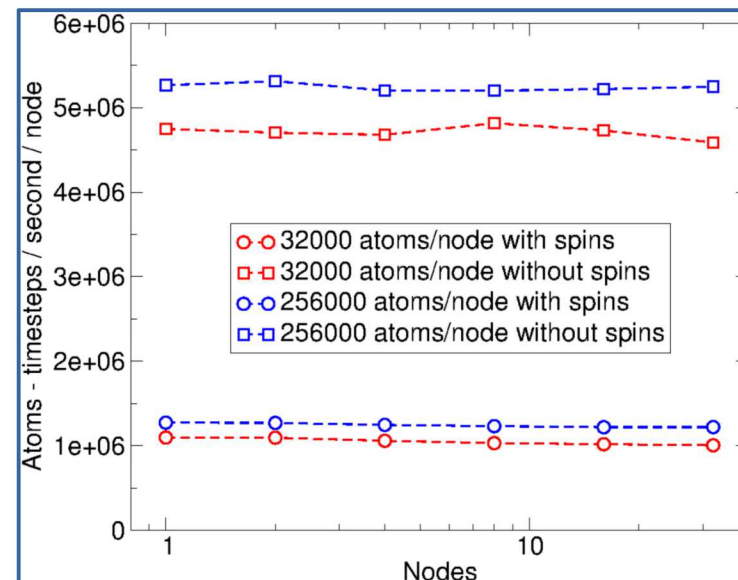
A scalable implementation of SD-MD in LAMMPS

► Accuracy of the parallel algorithm



- Error for the total energy and the norm of the magnetization preservation.
- Different trajectories, but no accuracy loss with the number of processes.

► Parallel efficiency: weak scaling



- From 1 to 32 Broadwell nodes, with 36 processes per node (1024 processes for the last point).
- Spin-lattice calculations only 5 times slower than an EAM calculation.



- Summary of the CS section:

- ▶ A symplectic scalable algorithm was implemented into LAMMPS
- ▶ This implementation tackles the two first limitations.

- Next section: Implemented physics, and examples

- ▶ Current interactions, and how to parametrize the exchange.
 - ▶ Two ongoing examples of applications.
- 

Accounting for the spin-lattice interactions

- The current version accounts for six magnetic interactions:

▶ Exchange interaction:

$$\mathbf{H}_{exchange} = - \sum_{i,j,i \neq j}^N J(r_{ij}) \vec{s}_i \cdot \vec{s}_j$$

- Simulation of ferromagnetism, antiferromagnetism, ferrimagnetism, ...

▶ Uniaxial anisotropy:

$$\mathbf{H}_{an} = - \sum_{i=1}^N K_{an}(\mathbf{r}_i) (\mathbf{s}_i \cdot \mathbf{n}_i)^2$$

- Simulation of magnetocrystalline anisotropy or shape anisotropy.
- ✗ Poor lattice dependence.

▶ Dzyaloshinskii-Moriya:

$$\mathbf{H}_{dm} = \sum_{i,j=1,i \neq j}^N \vec{D}(r_{ij}) \cdot (\vec{s}_i \times \vec{s}_j),$$

- Simulation of an effect of the spin-orbit coupling
- Very trendy (chiral magnetism, skyrmions...)

▶ Zeeman interaction:

$$\mathbf{H}_{Zeeman} = -\mu_B \mu_0 \sum_{i=0}^N g_i \mathbf{s}_i \cdot \mathbf{H}_{ext}$$

- Interaction with an external magnetic field (constant or time dependent).

▶ Magneto-electric interaction:

$$\mathbf{H}_{me} = - \sum_{i,j,i \neq j}^N \left(\vec{E} \times \vec{e}_{ij} \right) \cdot (\vec{s}_i \times \vec{s}_j),$$

- Interaction between spins and electric dipoles.
- Simulation of multiferroic materials.

▶ Néel pair anisotropy:

$$\mathbf{H}_{Néel} = - \sum_{i,j=1,i \neq j}^N g_1(r_{ij}) \left((\mathbf{e}_{ij} \cdot \mathbf{s}_i)(\mathbf{e}_{ij} \cdot \mathbf{s}_j) - \frac{\mathbf{s}_i \cdot \mathbf{s}_j}{3} \right)$$

- Other way to account for effects of the spin-orbit coupling.
- Simulation of magnetocrystalline anisotropy and magneto-elasticity.

Parametrization of the exchange interaction

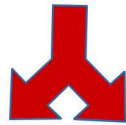
► Derivation of a force and a magnetic torque from the Hamiltonian:

$$\mathbf{H}_{exchange} = - \sum_{i,j,i \neq j}^N J(r_{ij}) \vec{s}_i \cdot \vec{s}_j$$



- Application of the following Poisson bracket:

$$\{f, g\} = \sum_{i=1}^N \left[\frac{\partial f}{\partial \mathbf{r}_i} \cdot \frac{\partial g}{\partial \mathbf{p}_i} - \frac{\partial f}{\partial \mathbf{p}_i} \cdot \frac{\partial g}{\partial \mathbf{r}_i} - \frac{\mathbf{s}_i}{\hbar} \cdot \left(\frac{\partial f}{\partial \mathbf{s}_i} \times \frac{\partial g}{\partial \mathbf{s}_i} \right) \right]$$



$$\vec{F}_i = \sum_j^{Neighbor} \frac{\partial J(r_{ij})}{\partial r_{ij}} (\vec{s}_i \cdot \vec{s}_j) \vec{r}_{ij}$$

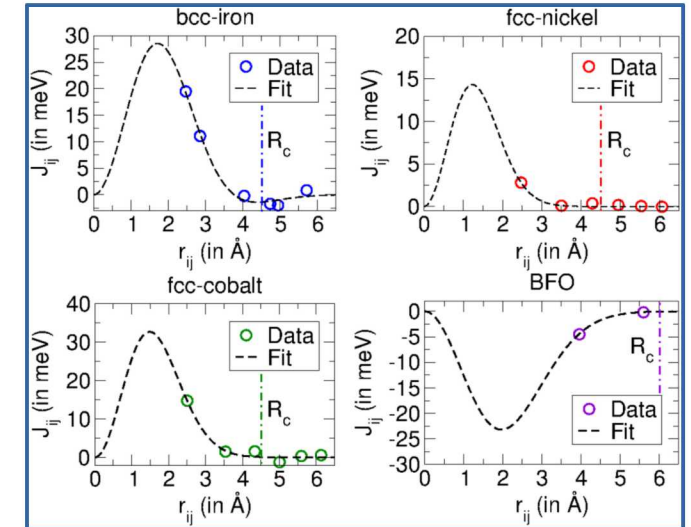
- Force applied to the atoms.

$$\vec{\omega}_i = \frac{1}{\hbar} \sum_j^{Neighbor} J(r_{ij}) \vec{s}_j$$

- Torque applied to the spins.

► Intensity of the coupling:

- Values of the exchange interaction from DFT calculations, or measurement:

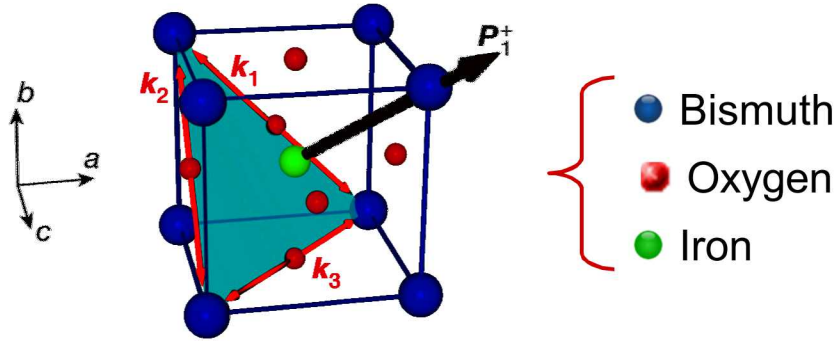


- Function used to fit the discrete values DFT results:

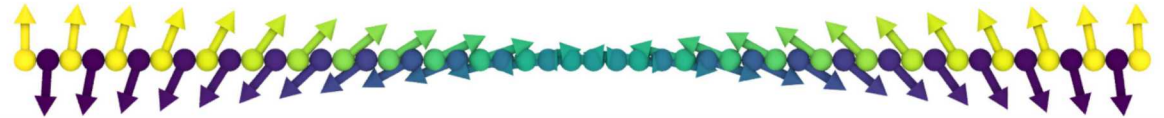
$$J(r_{ij}) = 4a \left(\frac{r_{ij}}{d} \right)^2 \left(1 - b \left(\frac{r_{ij}}{d} \right)^2 \right) e^{-\left(\frac{r_{ij}}{d} \right)^2} \Theta(R_c - r_{ij})$$

Example I: multiferroicity and spin textures in bismuth oxide BiFeO_3 (BFO)

3



► A 64 nm long cycloid appears due to the interplay between magnetic and ferroelectric orders:



► BFO Hamiltonian:

- We first only account for the iron atoms.
- The simulations are performed on a fixed lattice, using the following Hamiltonian:

$$\mathcal{H}_{\text{BFO}} = - \underbrace{\sum_{i,j,i \neq j}^N J(r_{ij}) \mathbf{s}_i \cdot \mathbf{s}_j}_{\text{AF exchange interaction}} + \underbrace{\sum_{i,j=1,i \neq j}^N (\mathbf{E} \times \mathbf{e}_{ij}) \cdot (\mathbf{s}_i \times \mathbf{s}_j)}_{\text{ME interaction}} - \underbrace{\sum_{i=1}^N K_{an}(r_i) (\mathbf{s}_i \cdot \mathbf{n}_i)^2}_{\text{Anisotropy}}$$

► Parameters:

- AF exchange interaction: $\begin{cases} J_1 = -4.38 \text{ meV} \\ J_2 = -0.2 \text{ meV} \end{cases}$

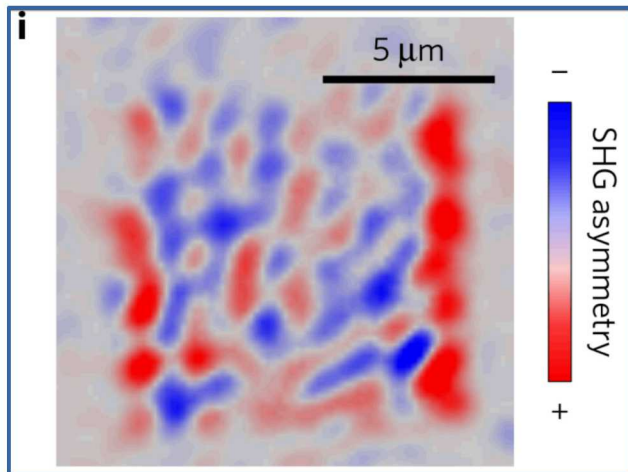
- Magneto-electric interaction:
 $E = 0.109 \text{ meV}$ along [111]

- Magnetic anisotropy:
 $K_{an} = 0.0033 \text{ meV}$ along [111]

Example I: multiferroicity and spin textures in bismuth oxide BiFeO_3 (BFO)

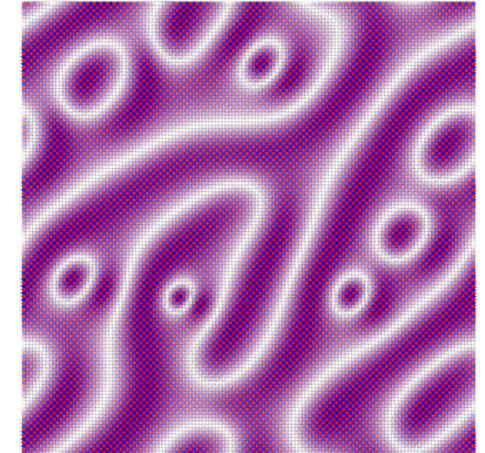
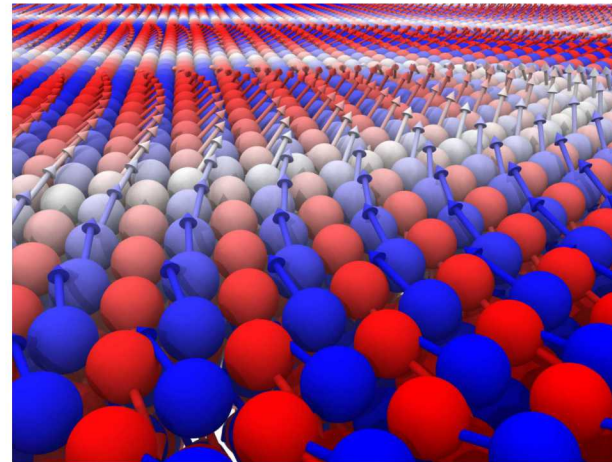
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► Experimental target:



► First numerical results:

► Equilibrium configuration after relaxation:



Chauleau, J. Y., Viret, M et al. (2017). Multi-stimuli manipulation of antiferromagnetic domains assessed by second-harmonic imaging. *Nature materials*, 16(8), 803.

◆ Perspectives:

- Phase diagram of the existence of the antiferromagnetic bubbles.
- Find the correct interactions to perform full magneto-electric and elastic simulations.

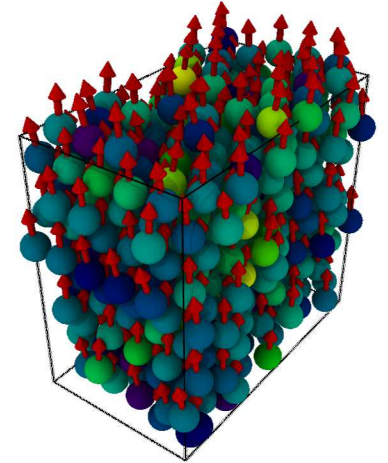
Example 2: magnetoelasticity of hcp cobalt

- Exchange interaction and EAM potential;

$$\mathcal{H}_{\text{hcp-Co}} = \sum_{i=1}^N \frac{|\mathbf{p}_i|^2}{2m_i} + \underbrace{\sum_{i,j=1}^N V(r_{ij})}_{\text{EAM potential} \Rightarrow \textcircled{1}} - \underbrace{\sum_{i,j,i \neq j}^N J(r_{ij}) \mathbf{s}_i \cdot \mathbf{s}_j}_{\text{Exchange interaction} \Rightarrow \textcircled{2}}$$

1 Pun, G. P. et al.. (2012). Phys. Rev. B, 86(13), 134116.

2 Sabiryanov, R. F. et al.. (1999). Phys. Rev. Lett., 83(10), 2062.



- Spin-orbit coupling and magneto-elastic effects with Néel dipolar and quadrupolar functions:

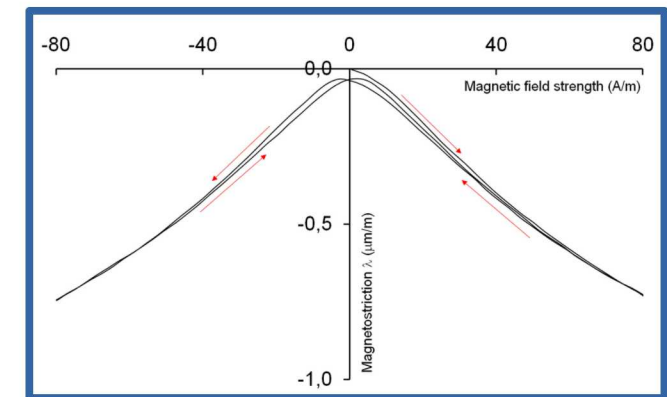
$$\mathcal{H}_{\text{Néel}} = - \sum_{i,j=1,i \neq j}^N g_1(r_{ij}) \left((\mathbf{e}_{ij} \cdot \mathbf{s}_i)(\mathbf{e}_{ij} \cdot \mathbf{s}_j) - \frac{\mathbf{s}_i \cdot \mathbf{s}_j}{3} \right) + q_1(r_{ij}) \left((\mathbf{e}_{ij} \cdot \mathbf{s}_i)^2 - \frac{\mathbf{s}_i \cdot \mathbf{s}_j}{3} \right) \left((\mathbf{e}_{ij} \cdot \mathbf{s}_i)^2 - \frac{\mathbf{s}_i \cdot \mathbf{s}_j}{3} \right) + q_2(r_{ij}) \left((\mathbf{e}_{ij} \cdot \mathbf{s}_i)(\mathbf{e}_{ij} \cdot \mathbf{s}_j)^3 + (\mathbf{e}_{ij} \cdot \mathbf{s}_j)(\mathbf{e}_{ij} \cdot \mathbf{s}_i)^3 \right)$$

Pair model from the magnetostriction constants

- Following Cullen's approach, magnetostriction constants can be expressed as functions of g_1 , q_1 , and q_2 .
- Knowing the experimental values of λ , one can fit the constant g_1 , q_1 and q_2 .
- Using spin-dependent DFT to check the obtained energy surface.

$$\left. \begin{aligned} \lambda_1^\alpha &= \frac{-B_1^\alpha [g_1(r_{ij}), q_1(r_{ij}), q_2(r_{ij})]}{C_1^\alpha} \\ \lambda_2^\alpha &= \frac{-B_2^\alpha [g_1(r_{ij}), q_1(r_{ij}), q_2(r_{ij})]}{C_2^\alpha} \\ \lambda^\epsilon &= \frac{-B^\epsilon [g_1(r_{ij}), q_1(r_{ij}), q_2(r_{ij})]}{C_{44}} \\ \lambda^\gamma &= \frac{-B^\gamma [g_1(r_{ij}), q_1(r_{ij}), q_2(r_{ij})]}{C_{11} - C_{22}} \end{aligned} \right\}$$

Target result



● Summary of the physics section:

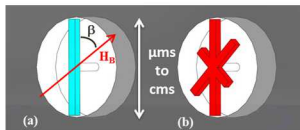
- ▶ The current implementation accounts for six interactions.
- ▶ The exchange interaction can be parametrized as a function of the interatomic distance.
- ▶ The simulation of BFO opens the door to coupled magneto-electric and elastic simulations.
- ▶ We started to work on the development of models accounting for the effects of the spin-orbit coupling.

Models of spin-orbit coupling

- ▶ Current model: pair Néel anisotropy, fitted for hcp cobalt:

$$\mathcal{H}_N = \sum_{i,j,i \neq j}^N g_1(r_{ij}) \left((\mathbf{e}_{ij} \cdot \mathbf{s}_i)(\mathbf{e}_{ij} \cdot \mathbf{s}_j) - \frac{1}{3} \mathbf{s}_i \cdot \mathbf{s}_j \right)$$

- ▶ Second approach: using spin-dependent DFT to fit g_1 , q_1 and q_2 , and check the obtained magnetostriction constants.
- ▶ Next steps: test this model and develop new ones for cubic crystals.
- ▶ Application to the simulation of magnetostriction in FeCo alloys for resonator devices.

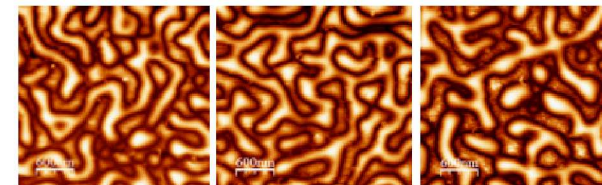


Single- (a) and multi-frequency (b) resonators

Long-range interaction

$$\mathcal{H}_{\text{Mag}} = - \underbrace{\sum_{i,j,i \neq j}^N J_{ij}(r_{ij}) \mathbf{s}_i \cdot \mathbf{s}_j}_{\text{exchange interaction}} - \underbrace{\frac{\mu_0 \mu_b^2}{4\pi} \sum_{i,j,i \neq j}^N \frac{g_i g_j}{r_{ij}^3} \left((\mathbf{s}_i \cdot \mathbf{e}_{ij})(\mathbf{s}_j \cdot \mathbf{e}_{ij}) - \frac{1}{3} (\mathbf{s}_i \cdot \mathbf{s}_j) \right)}_{\text{long-range dipolar interaction}}$$

- ▶ A direct calculation of this long-range Hamiltonian would scale as N^2
- ▶ We want to adapt a P3M to magnetic dipoles will be performed (scaling of $N \cdot \log(N)$).
- ▶ Application: simulation of the influence of a phase-change on the magnetic domain configuration.



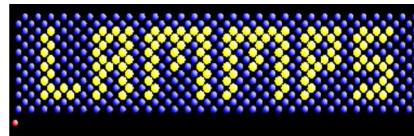
MFM measurements by T-M Lu, Sandia Labs

Conclusions

Conclusions:

- ◆ A massively parallel implementation of spin-lattice dynamics was implemented into LAMMPS (open release within the next months).
- ◆ The model adds new physics into LAMMPS, is accurate, scales very well with the number of processes, and only 5 times slower than usual MD – EAM calculations.
- ◆ Accounts for a lot of magnetic interactions, but models for the spin-orbit coupling and the long-range exchange interaction still need to be developed.
- ◆ Open to collaborations, feel free to contact us (jtranch@sandia.gov).

- Thanks to my group at Sandia:
Aidan Thompson, Steve Plimpton,
Mitch Wood, Stan Moore, Veena
Tikare.
- And to my Ph.D. advisor in
France:
Pascal Thibaudeau.



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Thank you your hosting me
and for you attention.