aC Weekly Report 1

Daniel Castillo Castro

April 23, 2020

Bulk Modulus in Lammps

Part of materials elastic properties (very underrated in academic teaching).

$$K = -V\frac{dP}{dV} \tag{1}$$

P=Pressure, V=Volume
It can be obtained by 2 ways:

- Using script in.elastic
- Calculating slope after expand and compress the sample.

Script method

- Script in.elastic is linked with the scripts:
  - init.mod: Read the initial sample (aC made by in.marks
  - potential.mod: Defines potential to use (EDIP in next results)
  - displace.mod: Define compression and expansion required for calculus.
- Pro: Gives data results faster and more automatically.
- Cons: The algorithm is more complex.

Slope Method

- Start expanding and compressing the sample using Lammps.
- Taking the slope of program evolution, $\frac{dp}{dV}$ is given.
  Multiplying by initial module it can be obtained K
- Pro: Easier implementation.
- Cons: More probability of human mistakes.

Compression and Expansion Process

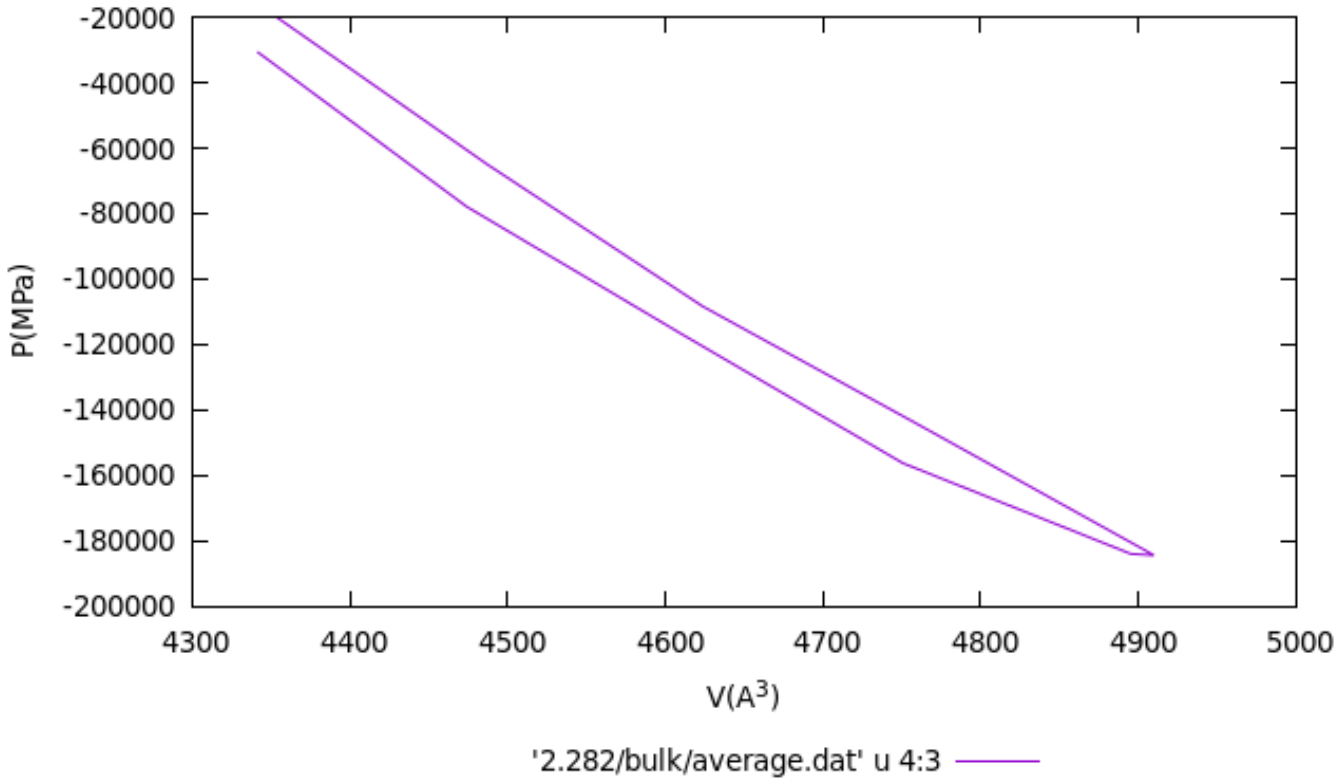

Figure: Process of Compression and Expansion of a sample of aC of initial density $\rho = 2.808 \frac{g}{cm^3}$, passing between volumes of $4900 A^3$ to $4300 A^3$. Used to find K via slope method.

First Running: Results

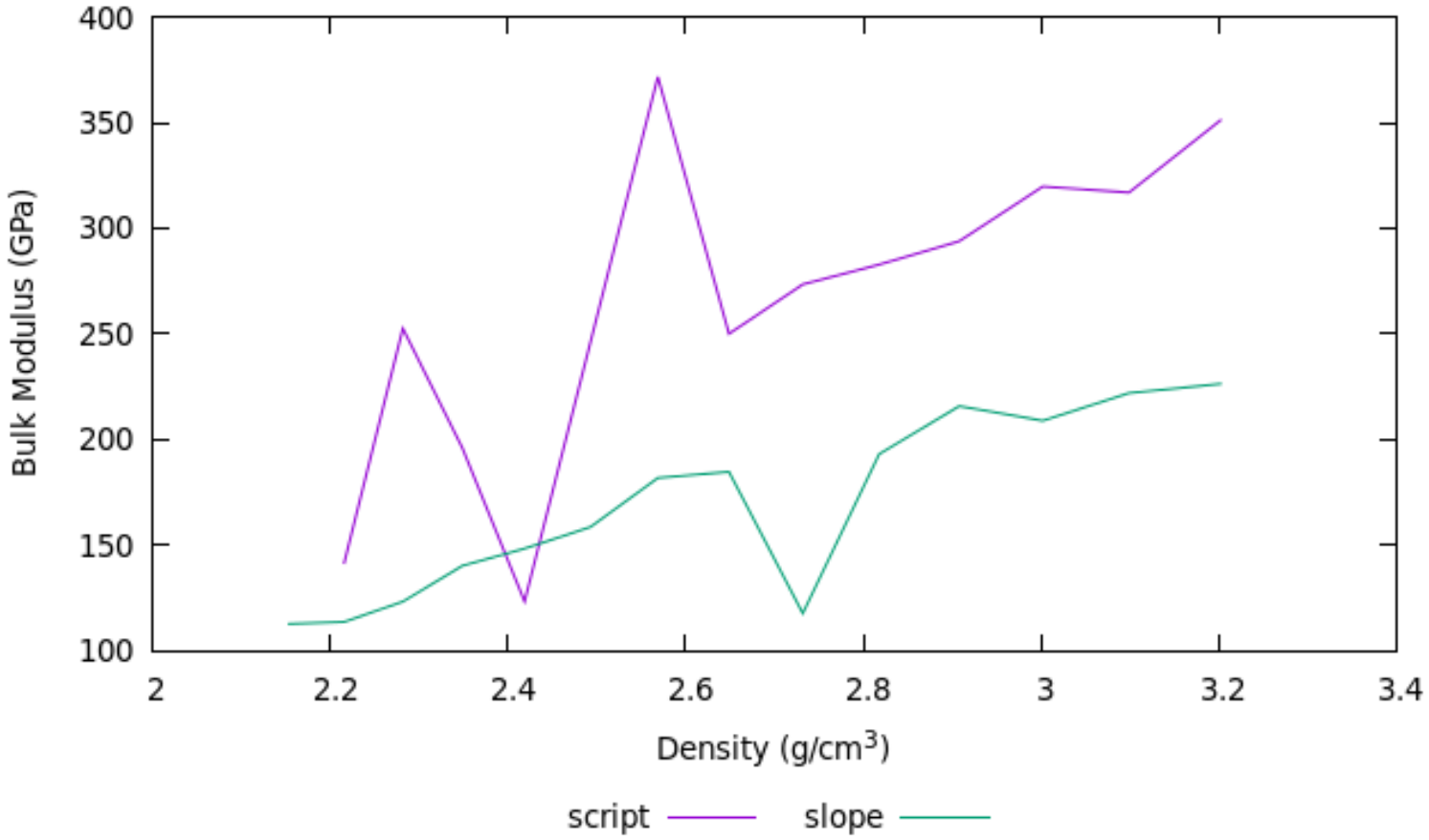

Figure: Bulk Modulus calculations using script method (pink line) and slope method (blue line). Great difference between the two results

aC Weekly Report 2

Daniel Castillo Castro

April 23, 2020

First Running: Analysis

- Why they are so different? $\Rightarrow$ 2 reasons:
  - The both methods needs to be modified (The evolución must be linear and not oscillating).
  - The metods works for differents temperatures (the script one at $T = 0K$, slope one at $T \sim 300K$).
- Possible Solutions
  - Correction to the both metods
  - The both methods must to work at $T \sim 0K$

Slope Method Corrections

- Improve data analysis considering

$$K = \frac{m * V}{0.0001 \frac{bar}{GPa}} \tag{2}$$

(m: curve slope in $\frac{atm}{A^3}$, V: volume in A^3)

- Change Temperature from $300K$ to $0K$ looking for results more similar to the script ones.
- Low compression and expansion rate from 5% to 1%, looking for a curve more linear.

Script Method Corrections

- Running it again (for consideration of random effects)
- Read input file and check its correctness
- It was observed 3 densities with incorrect data. It was proceed to correct it.

Corrected Compression and Expansion Process

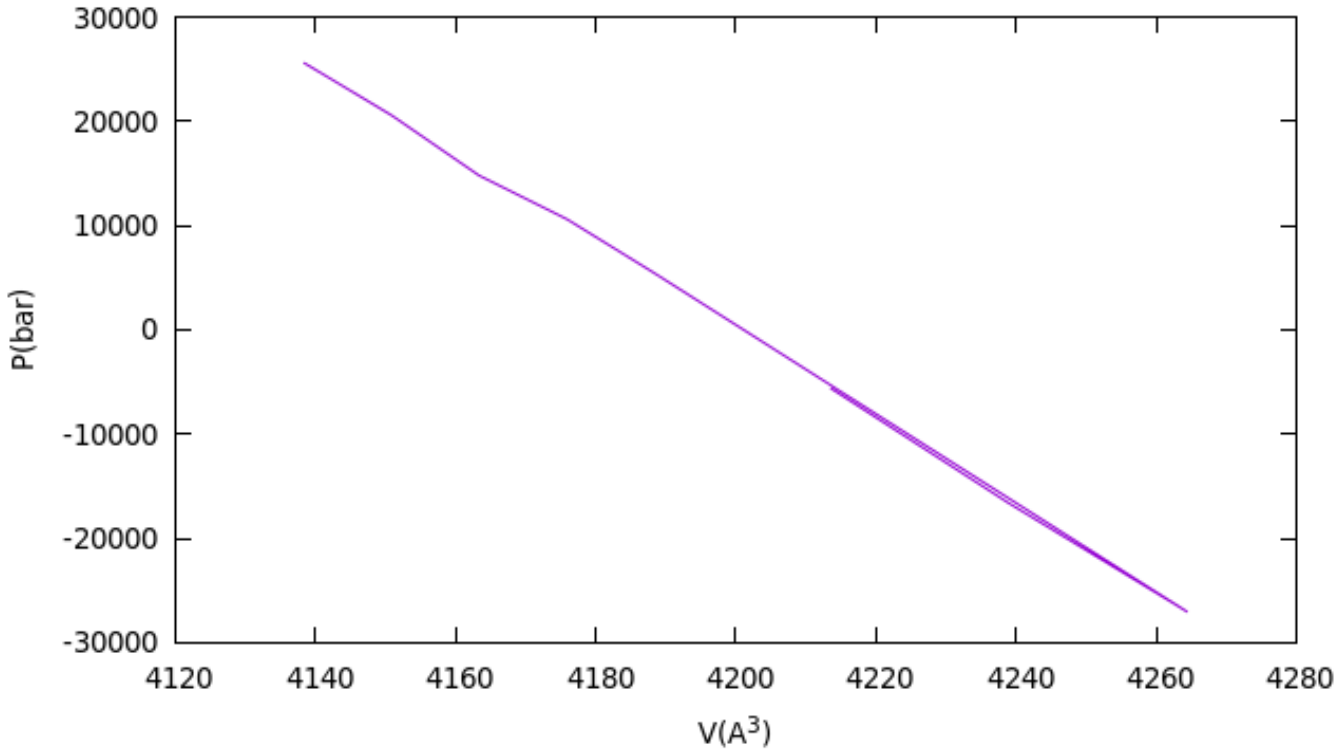

Figure: New Compression and Expansion process por the sample with $\rho = 2.808 \frac{g}{cm^3}$, going between $4120 \boldsymbol{A}^3$ to $4270 \boldsymbol{A}^3$. Used for slope method for $\boldsymbol{K}$ calculation.

Second Running: Results

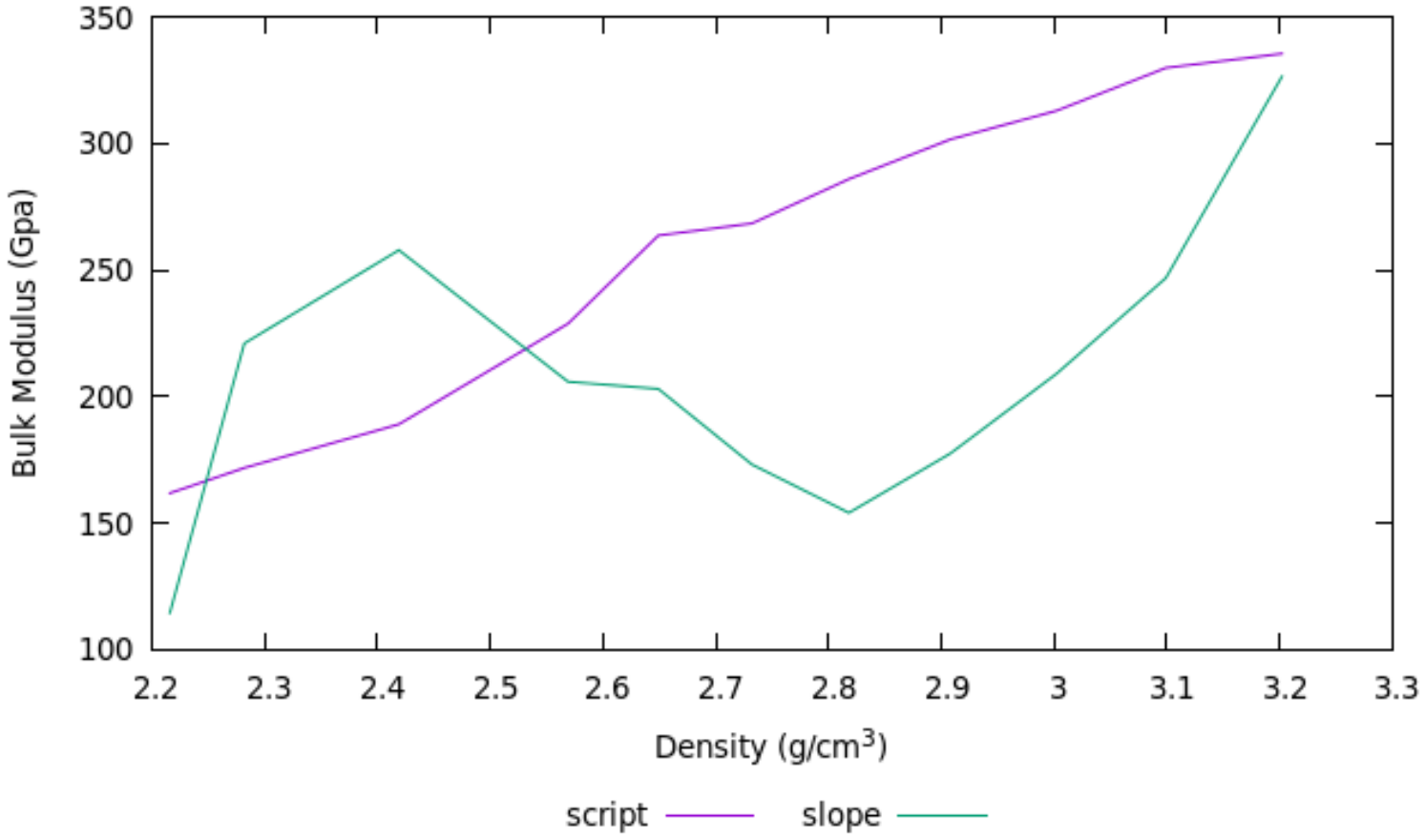

Figure: Bulk Modulus calculations using script method (pink line) and slope method (blue line). Results generated with 12 cores.

Second Running: Analysis

- Script Method obtains data that suggest a linear evolution (correct according to references)
- Slope Method doesn't. Even worse: Some result seems to be failed.
- Can de cores number affect the result? (With few the running could be slow, with too much could be clumpsy).
- What results will be obtained if core quantity is reduced? (from 12 to 8)

aC Weekly Report 3

Daniel Castillo Castro

April 23, 2020

Third Running, Results

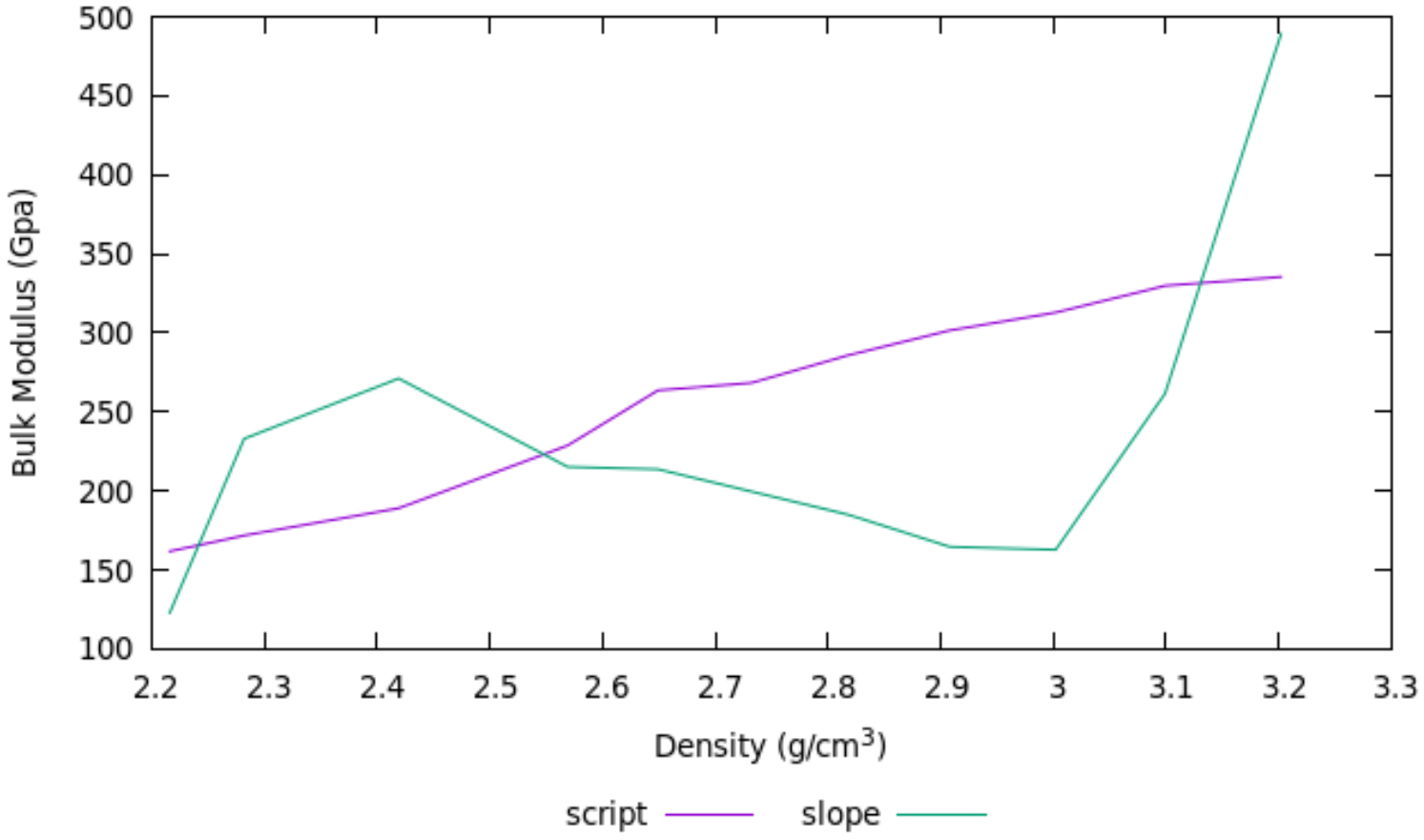

Figure: Bulk Modulus calculations using script method (pink line) and slope method (blue line). Results generated with 8 cores

Third Running, Analysis

- Data dispersion changes, but is not enough for the expectations.
- 1 possible option: lowering cores number even more (from 8 to 4)
- If this doesn't works, it will be needed another hypotesis.
- Effects of randomness of aC?

Correction for the Fourth Running

- Correction to density values in analysis because volume changes after initial relaxation in the both methods:

$$\rho = \frac{m}{V} = \frac{m}{V_0}\frac{V_0}{V} \tag{3}$$

(ρ: Density, m: Mass, V_0: Volume used before, V: Volume after relaxation)

- Give to script method mode time to relaxation process (it wasn't enough for reaching acceptable results).
- Use of XMgrace for fitting in slope method (Gnuplot fails).
- Improvement of data visualizacion of Bulk Modulus in Gnuplot.

Fourth Running: Results

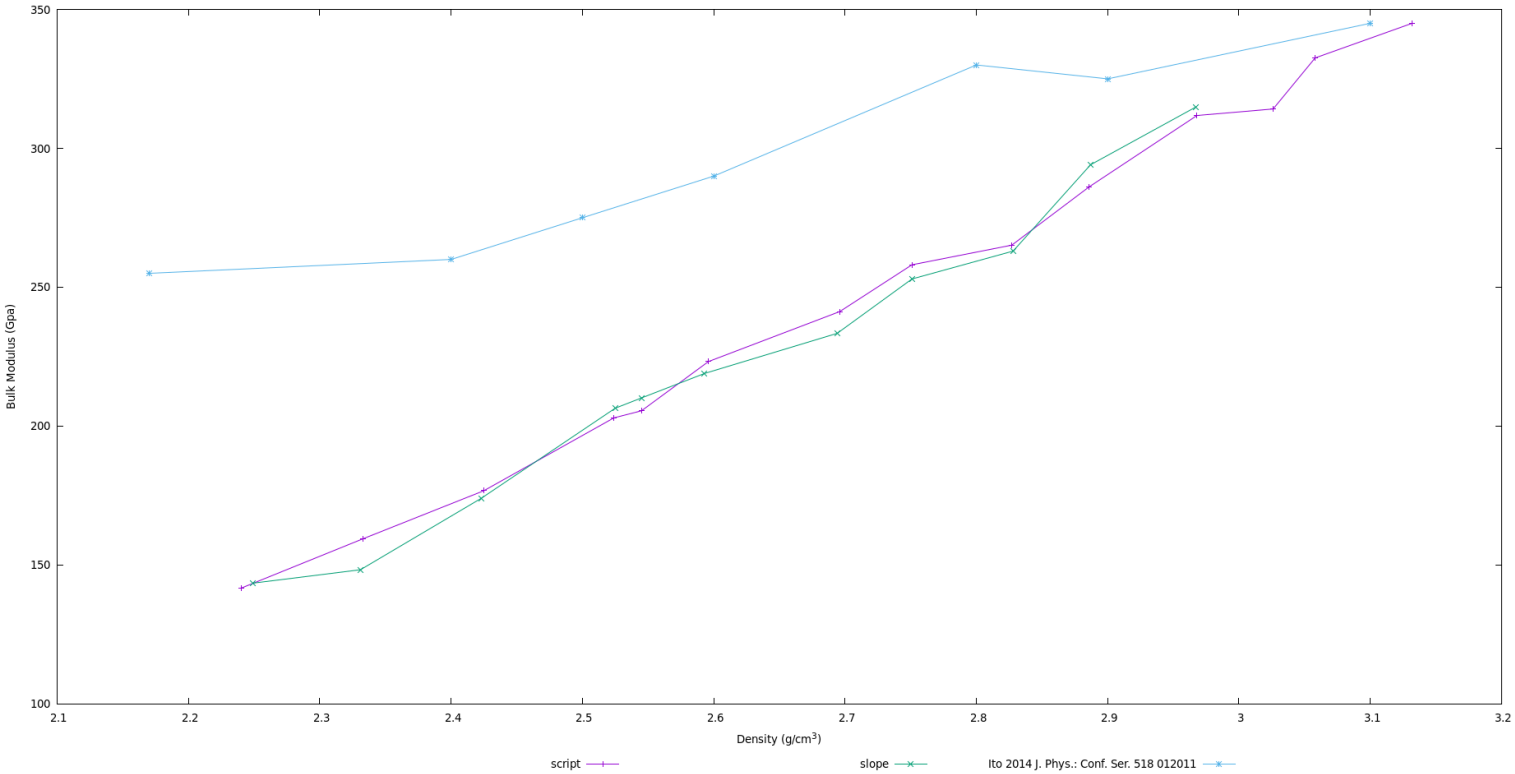

Figure: Bulk Modulus calculations using script method (pink line) and slope method (blue line). The both methods reach similar results.

Challenges from future research

- The both methods gives similar results, but there are lower than the results of Ito (Possible reason: it is a simulation with room temperature)
- There is a in.elastic script that includes variable temperature. Use it and tune it for the existing samples
- Compare the resuls of this script with Bulk Modulus calculations by slope method at room temperature ($\boldsymbol{T} = 300\boldsymbol{K}$)
- Look for and use another methods existing in Linux/Ubuntu for linear regression.

Projections of Research

- Make temperature ramps for all the existing ramps for search phase changes. Comparing it with Zazula.
- Using Ovito, checking out the coordination numbers for all the samples made.
- Using the elastic scripts, take the standard deviation of Young moduli in the 3 directions.
- Make several measurements for avoid random errors.

aC Weekly Report 4

Daniel Castillo Castro

April 23, 2020

in.elastic tuning

- The script for $\boldsymbol{T} \neq 0$ has more variables to tune for having an adequate result.
- Expected result: a linear increase of Bulk Moduli, a little bit greater than the $\boldsymbol{T} = 0$ case.
- The example script for the new case is found in folder lammps/tree/master/examples/ELASTIC_T.
- Taken the example, it was adapted for the case of the aC Lattice obtained before.

in.elastic tuning

- Variables tuned for the script:
  - up: Expansion of the material (it must be a very little number).
  - nrepeat: Number of samples (it can be any quantity for fitting).
  - nevery: Sampling interval (it can be greater).
- After one week of changing this variables and running the scripts again (for EDIP potential) it couldn't be obtained a satisfactory result.
- What change can make the script to generate correct results? It has be needed another kind of change?

in.elastic tuning

- Another variables that could be tuned:
  - timestep: Time used for every program run (Is enough time to take the result?)
  - tdamp: Time constant for thermostat (Can affect the result?)
  - neighbor: Can some weird neighbor behaviour set by the script damage the data?
- It was advised by Rafael that nonzero temperature script gives more errors to take the result.
- Is the results of room temperature fit with Ito's results?

New Slope Methods

- Linear Regression in Gnuplot fails. But why?
- Meanwhile, using XMGrace the results of Linear Regression seems more adequated.
- But XMGrace has a disadvantage: Is an old programm and can't run without GUI.
- Can I find a method via console that gives good results for Linear Regression?

New Slope Mehtods

- A possible candidate: Ubuntu commands like gmtgress or gblred
- Advantage: Looks ligther to run,, simpler to put in bin bash scripts.
- Disadvantage: Can give results with more errors (this is not a science base program).
- Can this kind of methods to be able to run datafiles like Lammps output files?

New Slope Methods

- Another candidate: A Python script.
- Libraries used: Pandas (data reading), Scikit (Realisation of Linear Regression Algorithm).
- Advantage: Can take datafiles using typical Python methods
- Disadvantage: It could be new errors related with arrange analysis.

aC Weekly Report 5

Daniel Castillo Castro

April 23, 2020

Development of To-Do list

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aC Weekly Report 6

Daniel Castillo Castro

April 23, 2020

Development of To-Do list

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aC Weekly Report 7

Daniel Castillo Castro

April 23, 2020

Development of To-Do list

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- blub

aC Weekly Report 8

Daniel Castillo Castro

April 23, 2020

Development of To-Do list

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- blub

NV Centers Weekly Report 1

Daniel Castillo Castro

April 23, 2020

NV Centers State of Art

- NV Center are very required for quibit representation.
- It can be possible to simulate and evaluate mechanical and vibrational properties using Classical Molecular Dynamics (LAMMPS).
- Is possible to add quantum effects in a Classical MD simulation, without using DFT?

4 possible NV Centers Hamiltonian

- Static Magnetic Field in $\hat{z}$

$$H = DS_Z^2 + \gamma_e B_z S_z \tag{4}$$

- Arbitrary Magnetic Field

$$H = DS_Z^2 + \gamma_e \vec{B} \cdot \vec{S} \tag{5}$$

- Static Magnetic Field in $\hat{z}$ and Nuclear Spin

$$H = DS_Z^2 + \gamma_e B_z S_z + \gamma_n B_z I_z + S_z A_{zz} I_z \tag{6}$$

- Static Magnetic Field in $\hat{z}$, Nuclear Spin and Anisotropy

$$H = DS_Z^2 + \gamma_e B_z S_z \gamma_n B_z I_z + S_z A_{zz} I_z + \frac{A_{ani}}{2} S_z (I_+ + I_-) \tag{7}$$

How to read the Hamiltonian?

- The electron spin of the defect in diamond is the qubit implementation.
- Electronic and Nuclear Spin can be affected by a Magnetic Field
- The dynamics given by the Hamiltonian explain the coherence and correlation of state in time evolution
- Trying to reproduce the phonon relaxation rates and energy levels of the system could be interesting.

Phonon Relaxation Rates

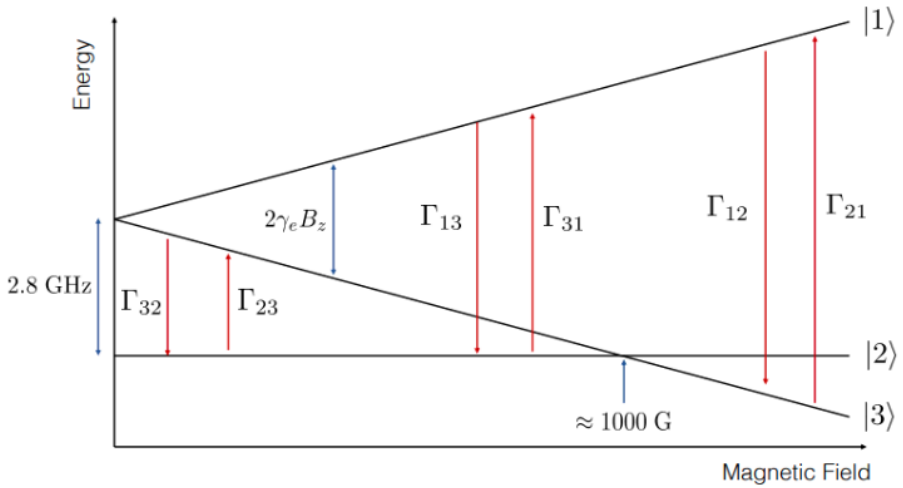

Figure 1: Phonon relaxation rates and energy levels of the ground triplet state of the NV^- center in diamond.

Figure: Representation of the reaction to an Spin Magnetic Field to an NV Center System. Taken from an internal script of Norambuena and Coto (in Dropbox Folder). This indicates that adding a external magnetic field to the MD system would be needed.

NV Centers MD Realisation.

Transactions of the Materials Research Society of Japan 36[1] 79-82(2011)

Molecular Dynamic Simulation for the NV-N Center Formation by Means of N_2 Beam Implantation into a Diamond

S. T. Nakagawa* [1], H. Kanda [2], T. Sakai [1], M. Ohishi [1], H. Saito [1], *S. Nakagawa [1], Y. Banden [1], and G. Betz [3]*

[1] *Graduate School of Science, Okayama Univ. of Science, Okayama 700-0005, JAPAN,*
Fax: +81 86 256 9458, e-mail:: stnak@dap.ous.ac.jp
[2] National Institute for Materials Science, 1-1 Namiki, Tsukuba-City, 305-0044, JAPAN,
[3] *Inst. für Allgemeine Physik, Technische Universität Wien, A-1040 Wien, AUSTRIA.*

- NV Center = change a carbon by nitrogen and remove another carbon in a neighbor.
- Change another carbon by nitrogen at relative distance $\Rightarrow$ NV-N Center (possible implementation of NOT gate).
- It was recomended to make a diamond lattice with the NV in center.

NV Centers MD Realisation.

- NV-N can be built bombarding with N atoms this lattice via Montecarlo Method
- Can be an easier form to put the second Nitrogen in Lammps?
- It would be needed the next libraries of Lammps:
  - spin: Makes Magnetic Spin Simulations. Allows to modify the magnetic field.
  - gcmc: Grand Canonical Monte Carlo simulation with interaction of an ideal gas (made of Nitrogen for our purposes).

Abinitio NV Centers Theory

DE GRUYTER Nanophotonics 2019; 8(11): 1907–1943

Review article

Ádám Gali*

***Ab initio* theory of the nitrogen-vacancy center in diamond**

- The point defect of the NV Center is Paramagnetic
- The eigenstates of the NV center can be labeled by C_{3v} symmetry irreducible representations (symmetry under rotation in $\frac{\pm 2\pi}{3}$ and 3 mirrors).
- It was required to see phonon interaction with the NV $\Rightarrow$ Phonolammps

Abinitio NV Centers Theory

- Abinitio simulation possible applications:
  - Interaction with another lattices and point defects.
  - Magnetooptical properties dependence of Thermodynamical changes (Temperature, Volume, Electromagnetic Field...)
- The second application was very few explored (!!!)
- This papers gives Fermi Energies that can be used as a reference for future results.

Abinitio NV Centers Examples

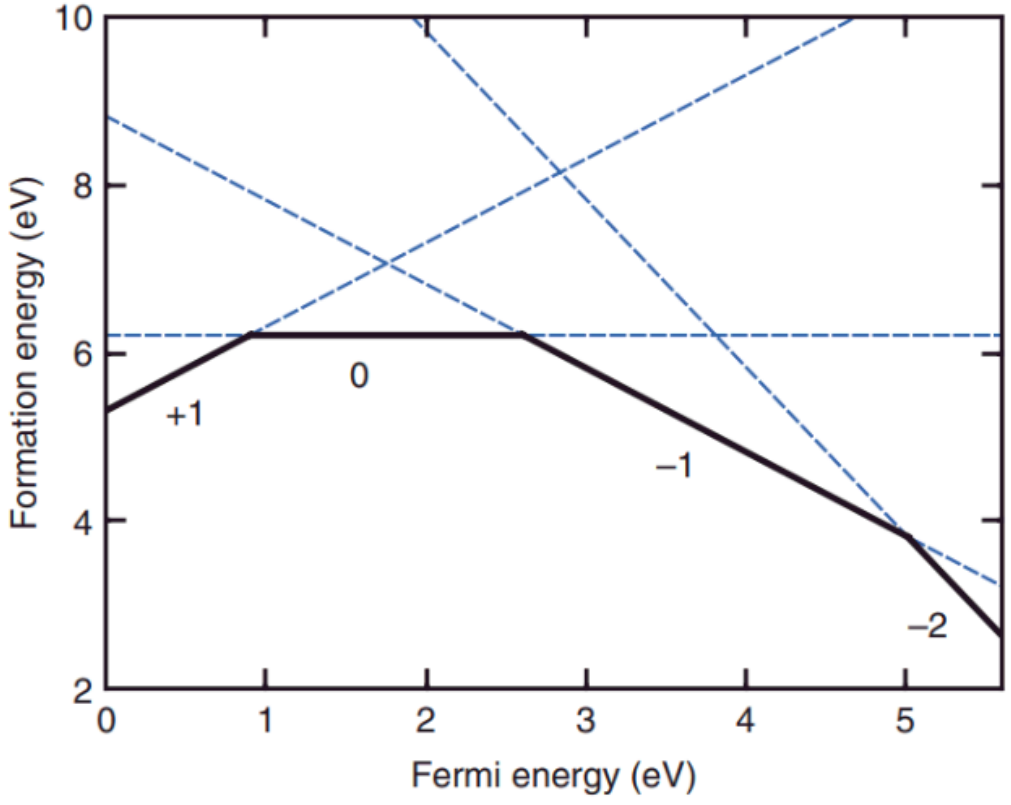

Figure 3: Formation energy of NV center in diamond [124]. The chemical potential of nitrogen was set to the energy of nitrogen atom in the nitrogen molecule at $T = 0$ K. The crossing lines correspond to the adiabatic ionization energies. The Fermi level is aligned to the top of the valence band. We note that these data differ, in particular, for the donor level from Ref. [131], which was obtained in a small 64-atom supercell.

NV Centers Weekly Report 2

Daniel Castillo Castro

April 23, 2020

First Principles Spin Calculation


npj | Computational Materials

www.nature.com/npjcompumats

REVIEW ARTICLE OPEN

First principles calculation of spin-related quantities for point defect qubit research

Viktor Ivády[1,2], Igor A. Abrikosov[1,3] and Adam Gali[2,4]

- Electronically, point defect system can be modeled as two level systems with a defined optical frecuency transition.
- Definition: ZPL: Zero Phonon Luminiscence = Difference between excited and ground state with zero phonons in the lattice.
- Another possible check calculation: Hyperfine tensor (result of electron-nucleus coupling) (!!!).

First Principles Bands Calculation

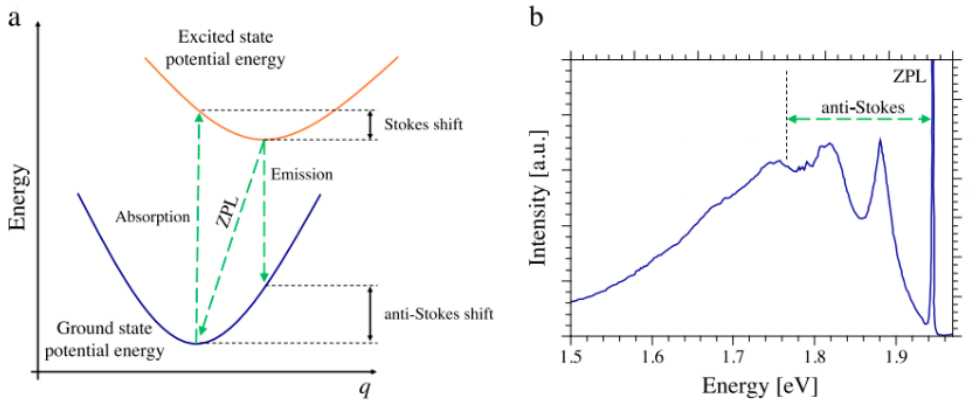

Fig. 3 **a** Schematic diagram of the configuration coordinate diagram of a point defect's ground and excited states. Green arrows show the most relevant phonon-assisted and zero-phonon (ZPL) optical transitions. **b** Luminescence spectrum of the NV center (data from ref. [158])

- ZPL can be found in MD finding the energies for zero photons.
- Another relevant concept: Luminiscence Spectrum, seems to be a ***fingerprint*** of the material.

Initial Project

1. Measure vibrational modes of diamond crystal simulations using con PhonoLAMMPS and comparing potentials Tersoff, ReboScr, ReaxFF y COMB.
2. Add vacancies, measure modes again and compare with DFT results.
3. Check for any potential if the results works (sequentially: ReaxFF, COMB, Tersoff).
4. Add nitrogen atoms to the system which gives the best results.
5. Obtain (Spectral Function) S_q and comparing it with experimental and DFT results.
6. Make the same procedure for nanoclusters, materials with defects, dislocations...

Thermodynamics and defect concentration

PHYSICAL REVIEW B **85**, 014102 (2012)

***Ab initio* thermodynamics calculation of the relative concentration of NV^- and NV^0 defects in diamond**

B. T. Webber,[1] M. C. Per,[1] D. W. Drumm,[2] L. C. L. Hollenberg,[2] and S. P. Russo[1]
[1]*Applied Physics, School of Applied Sciences, RMIT University, Melbourne, Victoria 3001, Australia*
[2]*School of Physics, University of Melbourne, Parkville, Victoria 3010, Australia*
(Received 10 July 2011; revised manuscript received 13 November 2011; published 5 January 2012)

- It can be formed ***NV^-*** and ***NV^0*** defects in a diamond lattice
- For the applications (Decoherence, ***QKD***, ***QI***) it is needed to have a greater concentration of ***NV^-***
- In Laboratory, NV Centers are made by ***thermal annealing***.

Thermodynamics and defect concentration

- DFFE: Defect Formation Free Energy

$$\Delta F_f^D = F^D - [F^H + \sum_d n_d \mu_d - \sum_h n_h \mu_h - q\mu_e] \qquad (8)$$

n_h: Host Atoms ,n_d: Defect Atoms, μ_d, μ_e, μ_h: Chemical Pot.

- Relative DFEE:

$$\Delta F_f^{Rel} = \Delta F_f^{NV^-} - \Delta F_f^{NV^0} = F^{NV^-} - F^{NV^0} + q\mu_e \qquad (9)$$

- If $\Delta F_f^{Rel} > 0$, the NV^0 centers are more stable thermodynamically.

Thermodynamics and defect concentration

- Equilibrium Defect Concentration: Directely related with DFFE

$$C^D(T) = n_I n_0 e^{-\beta \Delta F_f^D} \tag{10}$$

n_I: number of defects n_0: number of orientations by defect

- A charge from a reservoir system (a pure diamond bulk) is needed to make a bulk with a defined concentration of NV^-
- Relative Concentration: Possible quantity to label simulations.

$$C^{rel} = \frac{C^{NV^-}}{C^{NV^0}} = \frac{(n_I n_0)^{NV^-}}{(n_I n_0)^{NV^0}} e^{-\beta \Delta F_f^{Rel}} \tag{11}$$

NV Centers Weekly Report 3

Daniel Castillo Castro

April 23, 2020

Vibrational and Electronic Dynamics

nature physics

ARTICLES

PUBLISHED ONLINE: 29 SEPTEMBER 2013 | DOI: 10.1038/NPHYS2753

Vibrational and electronic dynamics of nitrogen–vacancy centres in diamond revealed by two-dimensional ultrafast spectroscopy

V. M. Huxter[1†], T. A. A. Oliver[1], D. Budker[2] and G. R. Fleming[1*]

- It can be found using the next models:
  - GW Approximation (GWA: Simplifying selfenergies.
  - Bethe–Salpeter Equation (BSE): Bound states of two particle system (aproximation of QFT).
  - Manybody Pertubation Theory (MBPT): Enough for optical excitation
- The main goal: To find the dynamics of the vibrational bath, useful for understanding of optical dephasing and another optical induced effects.(!!!)

Vibrational and Electronic Dynamics

- Questions that this can solve:
  - Is the vibrational and electronic dynamics of NV Centers an effectof such vibrational bath?
  - Can the Dynamics of the vibrational bath to provoke ultrafast decay?
- Experimentally, this dynamics can be obtained by ultrafast measurements, called 2D Electronic Spectroscopy (2DES).
- In DFT simulations it was observed vibrational and electronic relaxations, reaching to a definition of Absortion Spectra
- The second derivative of Absortion Spectra is called Vibronic Structure. This structure is like a ***fingerprint*** of the defect.

NV Centers Transmision Luminescence

First-principles theory of the luminescence lineshape for the triplet transition in diamond NV centres

Audrius Alkauskas[1], Bob B Buckley[2], David D Awschalom[2,3] and Chris G Van de Walle[1]

[1] Materials Department, University of California, Santa Barbara, California, USA
[2] Center for Spintronics and Quantum Computation, University of California, Santa Barbara, California, USA
[3] Institute for Molecular Engineering, University of Chicago, Chicago, Illinois, USA
E-mail: alkauskas@engineering.ucsb.edu and vandewalle@mrl.ucsb.edu

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doi:10.1088/1367-2630/16/7/073026

- Almost all the NV Centers applications are coming from the measurement of photoluminiscence between excited and ground states of de NV Center system as a function of another experimental variables.

NV Centers Transmision Luminescence

- At low temperature the luminiscence band is a straight line in ZPL.
- Alkausas obtained the luminiscence spectra for NV Centers using DFT. Can this spectra be obtained by MD? (!!!)
- The electron and the rest of the lattice form a Jahn–Teller $E \otimes e$ system, that can be defined roughly as a geometrical distortion of the bands of the system, reducing energy and symmetry.

NV Centers Transmision Luminescence

- The Luminiscence Spectra analysis is made considering the following approximations:
  - Normal modes that contributes to luminiscence are the bulk with defect ones, more that the beam ones.
  - Modes in excited electronic state are the same than the ground electronic state.
- The second approximation doesn't work at all for NV Centers. But it can be compared with experimental data to make corrections for the model.
- Luminiscence Spectra is related with Spectral Density. Finding one of them it can be derived the another:

$$\boldsymbol{S}(\hbar\omega) = \sum_{\boldsymbol{k}} \boldsymbol{S_k}\delta(\hbar\omega - \hbar\omega_{\boldsymbol{k}}) \tag{12}$$

Method for implementation of MD Simulation

Diamond samples realisation

1. Make diamond lattice samples in the potentials Tersoff, Rebo, ReaxFF y COMB.
2. Take the library gcmc for bombarding a copy of the sample and obtaining NV and NV-N Systems
3. Evaluate the stability of the all the samples in each potential (changes of phase, elastic variables).

Method for implementation of MD Simulation

Lattice Measurements

1. Find the both ***fingerprints***: Luminiscence Spectrum and Vibrionic Structure using the photon and PhonoLammps libraries.
2. Find the following properties: Hyperfine Tensor, Dynamics of Vibrational Bath.
3. Evaluate the dependance of the properties of variables like Temperature, Pressure and External Magnetic Field

NV Centers Weekly Report 4

Daniel Castillo Castro

April 23, 2020

Lammps: Fix Phonon Command

- The command fix:phonon depends of PHONON package (Lammps must be compiled with this package)
- This package allow to use a command that has a detailed command to take phonon dynamics.
- This phonon dynamics can be taken to do band diagram user another code that it will explained later.

Lammps: Fix Phonon Command

- Syntax of fix phonon command:
  fix 1 all phonon 10 5000 500000 GAMMA OUT nasr 100
- Elements of syntax
  - 10: Timelapse between Green function measurements.
  - 5000: Lapse between measurements when dynamical matrix is obtained.
  - 500000: Lapse between measurements (Waiting time).
  - GAMMA: Generate map info internally.
  - nasr 100: Number of iterations to enforce acoustic sum rule.

Lammps: Fix Phonon Command

- Files used in every sample:
  - in.dia: Generate a Diamond Bulk using any potential.
  - dia.out: Output file made by in.dia
  - in.in: Take dia.out and measure the phonon spectra for the lattice.
- If it has obtained the phonon spectra, Can this spectra be visualized?
- It was needed a special tool for it.

Phana: Band Diagrams from Lammps Output

- Phana is for ***Phonon Analyzer***, takes a dump file and converts it in a datafile ready for Gnuplot
- It is in Lammps folder, in lammps/tools/USER/PHONON
- It was needed BLAS and LAPACK library. This can be installed in Linux following ***Illinois***
- It was tested in my PC (2 cores) and repeated the installation in the COIC PC (12 cores).

Phana: Band Diagrams from Lammps Output

- Phana is used putting the command
  phana Out.bin.6000000 < in.disp
- Elements of Syntax:
  - Out.bin.6000000: Binary file result of in.in-
  - in.disp: File with instructions for Phana
- The Phana gives a file that, ran in Gnuplot, gives the band diagram.

Test: Theory of Bands for Diamond Bulk

- For the COMB, EDIP, aiREBO and Tersoff potentials:
  1. Modify script from aC module to give diamond structure thermodynamically stable.
  2. Run it as in.dia files, giving the dia.out required
  3. Follow the method shown in the last slides.
- The band diagram for diamond can be found on references, for comparison with simulation data
- If it has found error, analise and fix it.

NV Centers Weekly Report 5

Daniel Castillo Castro

April 23, 2020

Development of To-Do list

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NV Centers Weekly Report 6

Daniel Castillo Castro

April 23, 2020

Development of To-Do list

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NV Centers Weekly Report 7

Daniel Castillo Castro

April 23, 2020

Development of To-Do list

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NV Centers Weekly Report 8

Daniel Castillo Castro

April 23, 2020

Development of To-Do list

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