

Order parameters of crystals in LAMMPS

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Abstract

To identify atoms in a bicrystal cell based on the grain to which they belong, LAMMPS is equipped with a feature called order parameters, i.e. the `fix orient/fcc` command [1]. The command has been adapted for b.c.c crystals in my thesis. This document aims to describe the order parameter calculation in detail, including its implementation in LAMMPS for b.c.c and f.c.c crystals

1 Formalism

1.1 Nearest neighbours

An atom i in the bulk of b.c.c (or f.c.c) crystal A is expected to have 8 (or 12 for f.c.c) nearest neighbours j , each at a distance $a_T\sqrt{3}/2$ (or $a_T\sqrt{2}/2$ for f.c.c) from i where a_T is the lattice constant at temperature T . Let \mathbf{R}_{ij}^A be the reference list for grain A, i.e. the list of vector position of neighbours j relative to the position of i . The list \mathbf{R}_{ij}^A is called the .

In the principal Cartesian frame, i.e. a frame whose orthogonal axes (\mathbf{X} , \mathbf{Y} , \mathbf{Z}) are ([100], [010], [001]), the neighbour list \mathbf{R}_{ij}^A contains the following vectors $[u_jv_jw_j]$,

$$\text{b.c.c} : \mathbf{R}_{ij}^A = \left\{ \begin{array}{cccc} \left[\frac{a_T}{2} \frac{a_T}{2} \frac{a_T}{2} \right] & \left[\frac{a_T}{2} \frac{a_T}{2} \overline{\frac{a_T}{2}} \right] & \left[\frac{a_T}{2} \overline{\frac{a_T}{2}} \frac{a_T}{2} \right] & \left[\overline{\frac{a_T}{2}} \frac{a_T}{2} \frac{a_T}{2} \right] \\ \left[\overline{\frac{a_T}{2}} \frac{a_T}{2} \overline{\frac{a_T}{2}} \right] & \left[\frac{a_T}{2} \overline{\frac{a_T}{2}} \overline{\frac{a_T}{2}} \right] & \left[\frac{a_T}{2} \overline{\frac{a_T}{2}} \frac{a_T}{2} \right] & \left[\overline{\frac{a_T}{2}} \overline{\frac{a_T}{2}} \frac{a_T}{2} \right] \end{array} \right\} \quad (1)$$

$$\text{f.c.c} : \mathbf{R}_{ij}^A = \left\{ \begin{array}{cccc} \left[\frac{a_T}{2} \frac{a_T}{2} 0 \right] & \left[\frac{a_T}{2} \overline{\frac{a_T}{2}} 0 \right] & \left[\overline{\frac{a_T}{2}} \frac{a_T}{2} 0 \right] & \left[\frac{a_T}{2} \overline{\frac{a_T}{2}} 0 \right] \\ \left[\frac{a_T}{2} 0 \frac{a_T}{2} \right] & \left[\frac{a_T}{2} 0 \overline{\frac{a_T}{2}} \right] & \left[\overline{\frac{a_T}{2}} 0 \frac{a_T}{2} \right] & \left[\overline{\frac{a_T}{2}} 0 \overline{\frac{a_T}{2}} \right] \\ \left[0 \frac{a_T}{2} \frac{a_T}{2} \right] & \left[0 \frac{a_T}{2} \overline{\frac{a_T}{2}} \right] & \left[0 \overline{\frac{a_T}{2}} \frac{a_T}{2} \right] & \left[0 \overline{\frac{a_T}{2}} \overline{\frac{a_T}{2}} \right] \end{array} \right\} \quad (2)$$

where $j \in [1, 8]$ for b.c.c or $j \in [1, 12]$ for f.c.c.

In a non-standard frame, the orthogonal axes (\mathbf{X} , \mathbf{Y} , \mathbf{Z}) are ($[h_x k_x l_x]$, $[h_y k_y l_y]$, $[h_z k_z l_z]$). The position of nearest neighbours $[u_j v_j w_j]$ in \mathbf{R}_{ij}^A , see Eqs. (1) and (2), can be transformed into $[u'_j v'_j w'_j]$ according to the non-standard frame via

$$u'_j = \frac{[u_j v_j w_j] \bullet [h_x k_x l_x]}{\sqrt{h_x^2 + k_x^2 + l_x^2}}, \quad v'_j = \frac{[u_j v_j w_j] \bullet [h_y k_y l_y]}{\sqrt{h_y^2 + k_y^2 + l_y^2}}, \quad w'_j = \frac{[u_j v_j w_j] \bullet [h_z k_z l_z]}{\sqrt{h_z^2 + k_z^2 + l_z^2}} \quad (3)$$

for all j (8 for b.c.c and 12 for f.c.c). The transformation described by Eq. (3) is particularly useful when working with bicrystal cells. In a bicrystal cell, the orthogonal axes of grain A and B are (\mathbf{X}_A , \mathbf{Y}_A , \mathbf{Z}_A) and (\mathbf{X}_B , \mathbf{Y}_B , \mathbf{Z}_B), respectively. As will be clear later, the implementation of order parameter calculation requires two supplementary files which contain vector positions of nearest neighbours of atoms in grain A and B. Eq. (3) is thus relevant for such purpose.

1.2 Order parameter

An unscaled order parameter $\tilde{\zeta}_i^A$ can be calculated for all atoms i based on the difference between the actual position of neighbours j relative to i \mathbf{r}_{ij} and the reference positions documented in the list \mathbf{R}_{ij}^A . It is defined as

$$\tilde{\zeta}_i^A = \frac{1}{N_{actual}} \sum_{j=1}^{N_{actual}} \min \left(\mathbf{r}_{ij} - \mathbf{R}_{ij}^A \right) \quad (4)$$

where $N_{actual} \leq N_{1nn} = 8$. The min function means that the unscaled order parameter $\tilde{\zeta}_i^A$ is calculated by assigning to each actual j one of the vectors from the reference list \mathbf{R}_{ij}^A such that their interatomic distance is minimum. This means that if an atom i is in the bulk of crystal A, all $\mathbf{r}_{ij} = \mathbf{R}_{ij}^A$ and its unscaled order parameter will be $\tilde{\zeta}_i^A = 0$.

If there is another crystal B, its reference being \mathbf{R}_{ij}^B , and atoms in crystal A are measured

relative to crystal B, the unscaled order parameter is

$$\zeta^{AB} = \frac{1}{N_{1nn}} \sum_{j=1}^{N_{1nn}} \min(\mathbf{R}_{ij}^B - \mathbf{R}_{ij}^A) \quad (5)$$

Note that ζ^{AB} is not necessarily the maximum possible value for ζ_i^A because the term $\min(\mathbf{R}_{ij}^B - \mathbf{R}_{ij}^A)$ is not equal for each j , making ζ^{AB} the average of ζ_i^A . If an atom i is only surrounded by one first-nearest neighbour j and that j happens to be the furthest from any vectors in the reference list \mathbf{R}_{ij}^B , the resulting ζ_i^A will definitely be higher than ζ^{AB} .

In order to standardize the range of order parameter across different grain boundaries, the normalized order parameter η_i^A is defined, i.e.

$$\eta_i^A = \begin{cases} 0 & \text{if } \zeta_i^A < \zeta_{lo} = K_{lo}\zeta^{AB} \\ \frac{(\zeta_i^A - \zeta_{lo})}{(K_{hi} - K_{lo})\zeta^{AB}} & \text{if } \zeta_i^A \in [\zeta_{lo}, \zeta_{hi}] \\ 1 & \text{if } \zeta_i^A > \zeta_{hi} = K_{hi}\zeta^{AB} \end{cases} \quad (6)$$

where K_{lo} , K_{hi} are cutoff values chosen arbitrarily between 0 to 1. A sensitivity analysis of cutoff values was discussed in [1]; typically, they are set as 0.25 and 0.75 respectively.

1.3 Artificial driving force technique

The order parameter η_i^A can further be used to induce grain boundary migration via the artificial driving force (ADF) technique [1, 2]. In this technique, the potential energy of atoms belonging to one grain of a bicrystal is raised. The added energy drives the boundary migration, shrinking the volume of high-energy grain. The added energy of an atom i , U_i^{ADF} , varies with the atomic local environment, as represented by the order parameter η_i , via [1]

$$U_i^{\text{ADF}} = \frac{1}{2}U_{\text{max}}(1 - \cos(\pi\eta_i)) \quad (7)$$

where U_{max} is the maximum energy added into any individual atom. The choice of trigonometric function is purely arbitrary and the resulting mobilities have been verified to be insensitive of the choice [3].

Only the dynamics of few atoms are affected by such an energy increase because the extra force acting on each atom is calculated based on the energy gradient. A non-zero extra force is only experienced by boundary atoms and their neighbours. Mathematically, atoms i with $\eta_i \in (0, 1)$ or those surrounded by at least 1 other atom j with $\eta_j \in (0, 1)$ will experience an extra force of $\mathbf{F}_i^{\text{ADF}}$, given by:

$$\begin{aligned}
\mathbf{F}_i^{\text{ADF}} &= -\frac{\partial U_{\text{tot}}^{\text{ADF}}}{\partial \mathbf{r}_i} = -\frac{\partial}{\partial \mathbf{r}_i} \sum_i U_i^{\text{ADF}} \\
&= -\frac{\pi U_{\text{max}}}{2\Delta\eta N_{\text{actual}}} \left[\left(\sum_j^{N_{\text{actual}}} \frac{\boldsymbol{\delta}_{ij}}{|\boldsymbol{\delta}_{ij}|} \right) \cdot \sin(\pi\eta_i) + \sum_j^{N_{\text{actual}}} \left(-\frac{\boldsymbol{\delta}_{ji}}{|\boldsymbol{\delta}_{ji}|} \cdot \sin(\pi\eta_j) \right) \right] \quad (8)
\end{aligned}$$

where the following definition applies:

$$\begin{aligned}
\boldsymbol{\delta}_{ij} &= \mathbf{R}_{ij}^A - \mathbf{r}_{ij} \\
\boldsymbol{\delta}_{ji} &= \mathbf{R}_{ji}^A - \mathbf{r}_{ji}
\end{aligned}$$

and \mathbf{r}_{ij} is the actual location of atom j relative from atom i , also $\mathbf{r}_{ij} = -\mathbf{r}_{ji}$. Note that the net force due to the ADF technique is non-translating, i.e. $\sum_i \mathbf{F}_i^{\text{ADF}} = 0$ where the summation index i is over all solvent atoms.

2 LAMMPS implementation

2.1 Overview

There are 3 main commands in LAMMPS relevant for the order parameter and the ADF formulation:

1. The main `fix orient` command.
2. The associated `fix_modify`, relevant for energy calculation.
3. The thermodynamic output `f_ID` and the per-atom output `f_ID[1]` and `f_ID[2]`.

The description of each command is explained in detail in the following sub-sections.

2.2 The `fix orient` command

Syntax:

```
fix [ID] [grp] orient/bcc <nstats> <dir> <latp> <Umax> <Klo> <Khi>↔
    <file0.txt> <file1.txt>
```

Anatomy:

[ID] ID of the `fix` command

[grp] Group of atoms to which the `fix` is applied. Use `all` only if there is 1 type of atoms. If there are more than 1 type of atoms, e.g. a binary alloy, one must first create an independent `grp` that encompasses atoms that constitute the crystal (solvent, not solute). The existing `fix` command in LAMMPS will not see this difference. The adapted `fix orient/bcc` has also been modified to now consider the possibility of having multiple type of atoms.

`orient/bcc` The b.c.c version of `orient/fcc`

$\langle nstats \rangle$ Number of time-steps at which the statistics of the `fix` command is printed. Use 0 as a default.

$\langle dir \rangle$ The direction of the grain boundary migration. Use $\langle dir \rangle$ of 1 if you want to use `file0.txt` as the reference orientation. Use $\langle dir \rangle$ of 0 if you want to use `file1.txt` as the reference orientation.

$\langle latp \rangle$ The lattice parameter used to compute the cutoff distance. LAMMPS by default **defines** $r_cutoff_FCC = 1.57 * 0.5 * \langle latp \rangle * \sqrt{2}$. The use of 1.57 is arbitrary since what LAMMPS does is, after collecting all neighbours within `r_cutoff_FCC`, sorting them out and then storing only the closest 12. If this definition makes you uncomfortable, you can always 'rescale' the cutoff distance by modifying the $\langle latp \rangle \Rightarrow \langle latp \rangle / 1.57$. The same applies for `r_cutoff_BCC`. Here, the $r_cutoff_BCC = 1.57 * 0.5 * \langle latp \rangle * \sqrt{3}$ and only the closest 8 will be stored.

$\langle Umax \rangle$ The maximum energy added to an atom, see Eq. (7). The unit is eV for units `metal`.

$\langle K1o \rangle$ The low-threshold for the order parameter, see Eq. (6), where $\langle K1o \rangle = 0.25$ in [1].

$\langle K1i \rangle$ The high-threshold for the order parameter, see Eq. (6), where $\langle K1i \rangle = 0.75$ in [1].

$\langle file0.txt \rangle$ $\langle file1.txt \rangle$

Each file contains the vector positions (x, y, z) of neighbour j of atom i for grain A (`file0.txt`) and B (`file1.txt`). The coordinates (x, y, z) **must be** in the actual distance unit, e.g. Å. There are 4 lines in both files for b.c.c (6 for f.c.c) bicrystals, representing half of the total number of nearest neighbours in a perfect crystal. This is because the rest of nearest neighbours are symmetrical to the origin. In other

words, if (x_j, y_j, z_j) denotes one neighbour, there is another neighbour located at $(-x_j, -y_j, -z_j)$. To identify the proper list of nearest neighbour positions, the transformation described by Eq. (3) can be used.

2.3 The `fix_modify` command

Syntax:

```
fix_modify [ID] energy yes
```

Anatomy:

This command allows one to display the net potential energy from the ADF technique contribution to the system, $U_{\text{tot}}^{\text{ADF}}$. When this command is added to the input file, the total potential energy will increase by an amount of $U_{\text{tot}}^{\text{ADF}}$. To print out only the extra potential energy, $U_{\text{tot}}^{\text{ADF}}$, see `f_ID` below.

2.4 The outputs `f_ID`, `f_ID[1]` and `f_ID[2]`

Syntax:

- Thermodynamic output: `thermo_style custom f_ID`
- Per-atom output: `dump [ID] all custom <nstats> f_ID[1] f_ID[2]`

Anatomy:

- Thermodynamic output: when enabled, `f_ID` displays $U_{\text{tot}}^{\text{ADF}}$ defined in Eq. (8).
- Per-atom output: when enabled, `f_ID[1]` and `f_ID[2]` display the unscaled order parameter $\tilde{\zeta}_i$, see Eq. (4), and the normalized order parameter η_i , see Eq. (6).

Important note:

- LAMMPS does not include the per-atom ADF energy calculated from Eq. (7) into its `pe/atom` output. However, one can easily know this per-atom quantity simply by substituting `f_ID[2]` into Eq. (7).

References

- [1] K.G.F. Janssens, D. Olmsted, E.A. Holm, S.M. Foiles, S.J. Plimpton, P.M. Derlet, *Computing the mobility of grain boundaries*. *Nat. Mater.* **5**, 124 (2006). doi:10.1038/nmat1559
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