LDCode Manual

Joseph E. Turney
Department of Mechanical Engineering
Carnegie Mellon University
Pittsburgh, PA 15213

August 25, 2010

1 Overview

Capabilities The lattice dynamics program, *LDCode*, performs harmonic and anharmonic lattice dynamics calculations. To do so, the program requires that a material, interatomic potential, and structure be given (see Section 2). From various options in the input file (*i.e.* without modifying the source code), the program is capable of computing and outputting:

- energy minimization using steepest decent
- harmonic dispersion curves between two arbitrary wave vectors with an arbitrary number of points,
- harmonic frequencies, eigenvectors, and velocities for all allowed phonons in the first Brillouin zone,
- phonon density of states from all allowed phonons in the first Brillouin zone,
- anharmonic frequency shifts and linewidths,
- and thermal conductivities.

Executing To run the code simply run the executable with no arguments like *./LDCode.exe*. The program will read the necessary input from the file *LD_Input.txt* which must be in the same directory as the executable. A different input file can be passed as the first (and only) argument to the program (*ex. ./LDCode input_file*. Note that whenever 'wave vectors' are listed in output files the numbers given are actually integer coefficients, l, to the the reciprocal lattice vectors, $2\pi\mathbf{b}$, divided by the number of unit cells, N. Thus, the actual wave vector is $\boldsymbol{\kappa} = 2\pi(l_0\mathbf{b}_0/N_0 + l_1\mathbf{b}_1/N_1 + l_2\mathbf{b}_2/N_2)$. The values of b are output in the file *LDCode.log*.

Output files The code produces a number of output files which contain a variety of information and data as described below.

- *stdout* Detailed information about the calculation including current status and errors are output to the screen.

- *LDCode.log* Always produced. Provides detailed information about the calculation including current status and errors. This is a good place to start if something unexpected happens.

- *Structure.xyz* The atomic positions of all the atoms in the crystal in angstroms.

- *Opt_Struct.xyz* The optimized atomic positions of all the atoms in the unit cell found through energy minimization.

- *Dispersion.xls* Output from the dispersion relation calculation. The first column is the magnitude of the wave vector times the direct lattice vectors divided by 2π. The remaining columns are the phonon frequencies in rad/ps.

- *Frequency.txt* Frequencies for all phonons in the first Brillouin zone in rad/ps. Output begins with the wave vector with the largest x-, y-, and z-components, incrementing down, starting with the z-component. Note that symmetry operations may be used to reduce the total number of wave vectors considered.

- *Eigenvector.txt* Normalized eigenvectors for all phonons in the first Brillouin zone scaled by $\exp[\boldsymbol{\kappa} \cdot \mathbf{r}_b]$, where $\mathbf{r}_b$ is the position vector of the b^{th} atom in the unit cell. The eigenvector components are arranged in rows in the same order as the frequencies in the file *Frequency.txt*. The format is the real part then the imaginary part associated with the x-direction of atom one, next the real and imaginary parts for the y-direction, etc. The fact that the eigenvector associated with a particular wave vector and dispersion branch is equal to the complex conjugate of the eigenvector associated with the negative of the wave vector and the same dispersion branch is used to reduce the total number of eigenvectors printed.

- *Velocity.txt* The x-, y-, and z-components of the group velocities for all phonons in the first Brillouin zone in m/s. These are in the same order as the frequencies in the file *Frequency.txt*.

- *DOS.xls* Output from the density of states calculation. The first column is the frequency in rad/ps. The second column is the number density in unit of 1e18 phonons/m^3.

- *Symmetry.txt* Lists the wave vectors associated with each row of output in the files *Frequency.txt* and *Velocity.txt*. Useful for identifying the wave vectors associated with the output when symmetry operations are used.

- *Shift_Width.txt* Contains the results of the anharmonic lattice dynamics calculation for a quantum system. The first column is the harmonic frequencies in rad/ps in the same order as printed in *Frequency.txt*. The second and third columns are the frequencies shifts in rad/ps from the third and fourth order derivatives of the potential energy. The fourth column is the relaxation time in ps. The file name is appended with the iteration number.

- *Classical_Shift_Width.txt* The same as *Shift_Width.txt* for a classical system.

- *Conductivity.xls* Stores the predicted thermal conductivity in the three Cartesian directions along with some other statistics and information computed from the anharmonic LD results.

– *Data_Direction.xls* Compiled data (wave vectors, frequencies, velocities, frequency shifts, lifetimes) along any reciprocal space direction present in the system.

– *Full_BZ.xls* Compiled data (wave vectors, frequencies, velocities, frequency shifts, lifetimes) for all phonons in the first Brillouin zone.

2 Usage

The input file for the lattice dynamics code must be named *LD_Input.txt* or passed as an argument to the program call. The input data is organized into categories which are identified with keywords. Additional keywords are available within some categories. All keywords, including the category keywords are not case sensitive. There are seven valid deliminators that can be used in the input file: space, tab, return, comma, equal sign, and vertical bar. Comments can be added when preceded by '%'. Simple mathematical expressions are valid where numerical values are expected. The mathematical expressions can correctly interpret the characters '(', ')', '^', '*', '/', '+', and '-' as well as user defined variables but may not contain any deliminators. Standard order of operation applies. There are no known errors in the evaluation of the mathematical expressions however it is best to use only simple expressions and verify their correct evaluation in the file *LDCode.log*.

Each category keyword is listed below followed by a detailed list of all the valid keywords for each category. Some of the categories are interdependent, so in the input file, the categories should be defined in the same order that they are listed below. All quantities should be entered in standard SI units (kg, m, s, K, and combinations thereof). In the following tables, italics denotes literal keywords.

The categories *VARIABLES* and *MATERIAL* do not have additional keywords. Under the *VARIABLES* category, the user can define variables. Variables names must begin with '$', may contain letters, numbers, and the underscore, and are case sensitive. All variables must be assigned a value at the time of their (re)definition. The value should be given after the variable name (*exs.* '$a 12', '$a_2=$a^2'). Under the *MATERIAL* category, the user should define a unique identifier for each material directly followed by its mass. The identifier is case sensitive and may not contain any special characters or be a keyword in the *UNIT_CELL* category. Using the *SYMMETRY* category will reduce the number of phonon modes that are written to the output files *Frequency.txt*, *Eigenvector.txt*, and *Velocity.txt*. One keyword, *wave_vector*, exists in the *SYMMETRY* category. Passing *true* as an argument to *wave_vector* will cause the file *Symmetry.txt* to be written. The default value for *wave_vector* is *false*. The keywords for the other categories are given in the tables below.

3 Known Issues

- None

Table 1: Category keywords and their arguments.

Category Keyword	Expected Argument	Category Description
VARIABLES	none	Category allows the user to define variables.
MATERIAL	integer>0	Argument defines the number of unique materials. Category defines the materials present in the system.
LATTICE	integer (currently '3' is the only valid argument)	Argument defines the dimension of the system. Category defines the direct lattice.
UNIT_CELL	integer>0	Argument defines the number of atom in the unit cell. Category defines the unit cell.
SYMMETRY	list of the equivalent directions w/o spaces	Category defines the symmetry of the lattice by identifying which lattice vector directions are equivalent. Currently symmetry should only be used for simple cubic (*123*) or rectangular lattices (*12*, *13*, or *23*). Defaults to no equivalent lattice vectors (same as entering *1*, *2*, or *3*). The symmetry $\boldsymbol{\kappa} = -\boldsymbol{\kappa}$ is general and is always used.
POTENTIAL	potential_keyword, integer>0, material identifiers	Arguments define the potential type, number of unique materials involved in the interactions, and a list of the material identifiers. Category defines the potential interactions. A separate potential is defined for each interaction.
PREPROCESS	*true* or *false*	Argument is a flag that specifies whether to perform certain pre-processing computations (*true*) or not (*false*). Setting it to *false* overrides keywords. Default is *false*.
DISPERSION	integer≥0	Argument defines the number of wave vectors used to compute the dispersion relation. Category defines the desired dispersion relation options.
HARMONIC	*true* or *false*	Argument is a flag that specifies whether to perform harmonic LD computations for the full BZ (*true*) or not (*false*). Setting it to *false* overrides keywords. Default is *true*.
ANHARMONIC	integer≥0	Argument defines the number of anharmonic iterations to perform. Defaults to zero. Category defines the anharmonic calculation options.
POSTPROCESS	*true* or *false*	Argument is a flag that specifies whether to perform certain post-processing tasks (*true*) or not (*false*). Setting it to *false* overrides keywords. Default is *false*.

Table 2: Keywords for the *LATTICE* category.

Keyword	Expected Argument	Description
a1 (*a2*, *a3*)	value	Direct lattice vectors defined in Cartesian coordinates.
bc1 (*bc2*, *bc3*)	*periodic*, *scattering*, or *free*	Boundary conditions along each lattice vector. *Scattering* is treated as *periodic* with an additional scattering mechanisms defined as twice the thickness divided by the group velocity.
N1 (*N2*, *N3*)	integer>0	Number of unit cells along each lattice vector. If the boundary condition is free the number of unit cells must be one.

Table 3: Keywords for the *UNIT_CELL* category.

Keyword	Expected Argument(s)	Description
coordinate	*Cartesian*, *direct*, or *reciprocal*	Defines coordinate system to use for input of atomic positions. *Direct* and *reciprocal* are along the direct and reciprocal lattice vectors. Can be redefined as many times as needed. *Cartesian* is default.
material identifier	3 values	Coordinates of atom. Done for each atom in the unit cell. The material identifiers are those defined under the *MATERIAL* category.

Table 4: Keywords for the *POTENTIAL* category. See Appendices A and B for additional information.

Keyword	Expected Argument	Description
cutoff	value(s)>=0	Cutoff distance in meters used for all but the anharmonic calculation.
Acutoff	value(s)>=0	Cutoff distance in meters used for the anharmonic calculation.
Other parameters	value(s)	Additional parameters are defined for individual potentials (see Appendix A).

Table 5: Keywords for the *PREPROCESS* category.

Keyword	Expected Argument	Description
optimization	integer	Specifies maximum number of iterations to perform to optimize the structure through a steepest decent energy minimization. A zero (default) or negative value suppresses the optimization routine.
structure	*true* or *false*	Flag to specify whether to output the initial atomic structure to the file *Structure.xyz* (*true*) or not (*false*). Default is *false*.
energy_force	*true* or *false*	Flag to specify whether to compute the energy and forces of/on the atoms and output them to the file *LDCode.log* (*true*) or not (*false*). Default is *false*.

Table 6: Keywords for the *DISPERSION* category.

Keyword	Expected Argument	Description
coordinate	*Cartesian*, *direct*, or *reciprocal*	Defines coordinate system to use for input of *begin* and *end*. *Direct* and *reciprocal* are along the direct and reciprocal lattice vectors. Can be redefined as many times as needed. *Cartesian* is default.
begin	3 values	Beginning wave vector for the dispersion curve. Does not include 2π.
end	3 values	Ending wave vector for the dispersion curve. Does not include 2π.

Table 7: Keywords for the *HARMONIC* category.

Keyword	Expected Argument	Description
frequency	*true* or *false*	Flag to specify whether to compute and output the phonon frequencies (*true*) or not (*false*). Default is *true*.
velocity	*true* or *false*	Flag to specify whether to compute and output the phonon velocities (*true*) or not (*false*). Default is *true*.
evect	*true* or *false*	Flag to specify whether to compute and output the phonon eigenvectors (*true*) or not (*false*). Default is *true*.
DOS	*true* or *false*	Flag to specify whether to compute and output the phonon density of states (*true*) or not (*false*). Default is *true*.

Table 8: Keywords for the *ANHARMONIC* category.

Keyword	Expected Argument	Description
temperature	value>0	System temperature.
fs_guess	value>0	Initial guess for the frequency shift as a fraction of the harmonic frequency. Outputs guesses to files *Shift_Width0.txt* and *Classical_Shift_Width0.txt*. Default is 0.005.
lw_guess	value>0	Initial guess for the linewidth as a fraction of the harmonic frequency. Outputs guesses to files *Shift_Width0.txt* and *Classical_Shift_Width0.txt*. Default is 0.005.
Q_interpolate	filename	File containing initial guesses for the quantum anharmonic calculation. Program will interpolate guesses for frequencies not in the list and writes to *Shift_Width0.txt*. The output from the anharmonic calculation can be used directly.
C_interpolate	filename	Same as Q_interpolate for the classical calculation and writes to *Classical_Shift_Width0.txt*.
conductivity	*true* or *false*	Flag to perform a thermal conductivity prediction or not. Default is *true*.
continue	integer≥0	Enter the value of the last iteration to continue the anharmonic calculation from that point. The program will determine if the files *Shift_Width.txt* and *Classical_Shift_Width.txt* are complete for the current iteration and pickup where it left off. This option is useful if an error occurred during an iteration or too few iterations were initially chosen to achieve convergence.

Table 9: Keywords for the *POSTPROCESS* category.

Keyword	Expected Argument	Description
TC_beg	integer≥0	Beginning anharmonic iteration number for which the thermal conductivity should be computed. This is not needed if the keyword *conductivity* in the *ANHARMONIC* category is set to *true*. A value of zero suppresses the thermal conductivity calculation during post-processing.
TC_end	integer≥0	Ending anharmonic iteration number for which the thermal conductivity should be computed. This is not needed if the keyword *conductivity* in the *ANHARMONIC* category is set to *true*. A value of zero suppresses the thermal conductivity calculation during post-processing.
data_itr	integer>0	Anharmonic iteration number for which data should be compiled (options controlled by the following keywords).
data_beg	3 integers	The integer coefficients l_0, l_1, and l_2 that specify a wave vector (see Section 1, paragraph on executing) at a starting position along an arbitrary direction. Harmonic and anharmonic phonon properties are compiled at this wave vector and output to the file *Data_Direction.xls*.
data_inc	3 integers	The integer coefficients l_0, l_1, and l_2 that specify a wave vector (see Section 1, paragraph on executing) by which to increment the starting position wave vector.
data_num	integer	The number of times to add the wave vector *data_inc* to the wave vector *data_beg*. Harmonic and anharmonic phonon properties are compiled at each of these wave vectors and output to the file *Data_Direction.xls*. In this way, the phonon properties along an arbitrary direction can be compiled and written. Set to a negative number (default) to suppress output.
data_fBZ	*no_symmetry*, *full_symmetry*, or *none*	This keyword controls the compilation of all the phonon property data in the first Brillouin zone and its output to the file *Data_fullBZ.xls*. The default flag is *none* which suppresses output. The flag *full_symmetry* uses the symmetry set in the *SYMMETRY* category to reduce the number of phonon modes output while *no_symmetry* does not apply any symmetry operations.

4 Possible Future Improvements

- Change the symmetry so that it is only computed once and store all needed data in arrays.
- Change potential energy storage to only have one instance of each potential used.
- Change potential energy calculation to compute total energy for a given potential.

A Interatomic Potential Definitions

Here, the interatomic potentials implemented in the LD code are described.

Lennard-Jones Potential identifier: *LJ*
Definition: $\Phi = \frac{1}{2}\sum_i \sum_j 4\epsilon_{ij}\left[\left(\frac{\sigma_{ij}}{r_{ij}}\right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}}\right)^{6}\right]$
Parameters: *cutoff*, *Acutoff*, ϵ, σ

Buckingham Potential identifier: *BUCK*
Definition: $\Phi = \frac{1}{2}\sum_i \sum_j A\exp\left[-\rho_{ij} r_{ij}\right] - \frac{c_{ij}}{r_{ij}^6}$
Parameters: *cutoff*, *Acutoff*, A, ρ

Coulomb Potential identifier: *COULOMB*
Definition: $\Phi = \frac{1}{4\pi\epsilon_0}\sum_i \sum_j \frac{q_i q_j}{r_{ij}}$
Parameters: *cutoff*, *Acutoff*, q, α
Notes: The charges are given in coulombs and are listed in the order of the list of unique materials involved in Coulomb interactions. The parameter α is used in the Wolf method for summing charges.

Stillinger-Webber: 2-body terms Potential identifier: *SW2*
Definition: $\Phi = \frac{1}{2}\sum_i \sum_j \epsilon_{ij} A_{ij}\left(B_{ij}\tilde{r}_{ij}^{-p_{ij}} - \tilde{r}_{ij}^{-q_{ij}}\right)\exp\left[1/(\tilde{r}_{ij} - a_{ij})\right]$ where $\tilde{r}_{ij} = r_{ij}/\sigma_{ij}$.
Parameters: a, ϵ, σ, A, B, p, q
Notes: Combine with the 3-body part of the Stillinger-Weber potential. The keywords *cutoff* and *Acutoff* are replaced by a in this potential.

Stillinger-Webber: 3-body terms Potential identifier: *SW3*
Definition: $\Phi = \frac{1}{6}\sum_i \sum_j \sum_k \left[h(\tilde{r}_{ij}, \tilde{r}_{ik}, \theta_{jik}) + h(\tilde{r}_{ji}, \tilde{r}_{jk}, \theta_{ijk}) + h(\tilde{r}_{ki}, \tilde{r}_{kj}, \theta_{ikj})\right]$ where $h(\tilde{r}_{ij}, \tilde{r}_{ik}, \theta_{jik}) = \epsilon_{jik}\lambda_{jik}\exp\left[\gamma_{ij}/(\tilde{r}_{ij} - a_{ij}) + \gamma_{ik}/(\tilde{r}_{ik} - a_{ik})\right]\left[\cos(\theta_{jik}) + 1/3\right]^2$ where $\tilde{r}_{ij} = r_{ij}/\sigma_{ij}$, $\tilde{r}_{ij} = r_{ij}/\sigma_{ij}$, and θ_{jik} is the angle formed by the vectors pointing from atom i to atom j and from atom i to atom k.
Parameters: a, ϵ, σ, λ, γ
Notes: Combine with the 2-body part of the Stillinger-Weber potential. The keywords *cutoff* and *Acutoff* are replaced by a in this potential.

REBO potential Potential identifier: *REBO*

Definition: See John Thomas, *Water flow and thermal transport through carbon nanotubes*. PhD Thesis, Carnegie Mellon University, Pittsburgh, PA (2010). Terms involving the dihedral angle are not implemented in the LD program.

Parameters: Same as used in John Thomas, *Water flow and thermal transport through carbon nanotubes*. PhD Thesis, Carnegie Mellon University, Pittsburgh, PA (2010).

Notes: The REBO potential is implemented as described above. This is not the full REBO potential. As currently implemented, there can only be one atom type interacting via the REBO potential and each atom must have exactly three neighbors.

B Potential Parameters for Multiples Species

When multiple species interact the potential parameters must be input in a particular format. The arguments expected immediately after the *POTENTIAL* keyword are the potential keyword (*ex. LJ* for Lennard-Jones), an integer giving the number of unique materials described by that potential, and a list of identifiers for those materials that interact with that potential. The order of the material identifiers in the list of unique materials is important to note when assigning the potential parameters. If the list of identifiers is: A, B, C, ... then for a two-body parameter, ϵ, the values are assigned in the order: ϵ_{AA}, ϵ_{AB}, ϵ_{AC}, ..., ϵ_{BB}, ϵ_{BC}, ..., ϵ_{CC}, etc. For a three-body parameter, λ, the order is: λ_{AAA}, λ_{AAB}, λ_{AAC}, ..., λ_{ABB}, λ_{ABC}, ..., λ_{ACC}, ..., λ_{BAA}, λ_{BAB}, λ_{BAC}, ..., λ_{BBB}, λ_{BBC}, ..., λ_{BCC}, ..., λ_{CAA}, λ_{CAB}, λ_{CAC}, ..., λ_{CBB}, λ_{CBC}, ..., λ_{CCC}, ..., etc. The first material listed in the subscript is assumed to be at the vertex of the angle formed by the triplet (if applicable). This behavior for ordering the potential parameters can be over-ridden in the code for special cases.