## Supplementary Information A:

### Discussion of running extended relaxation simulations

We present the results (*as plots*) of different thermodynamic variables for a sample study to highlight the importance for running long relaxation simulations (*specifically first NVE simulation for 15 ns*) prior to thermal transport simulations.

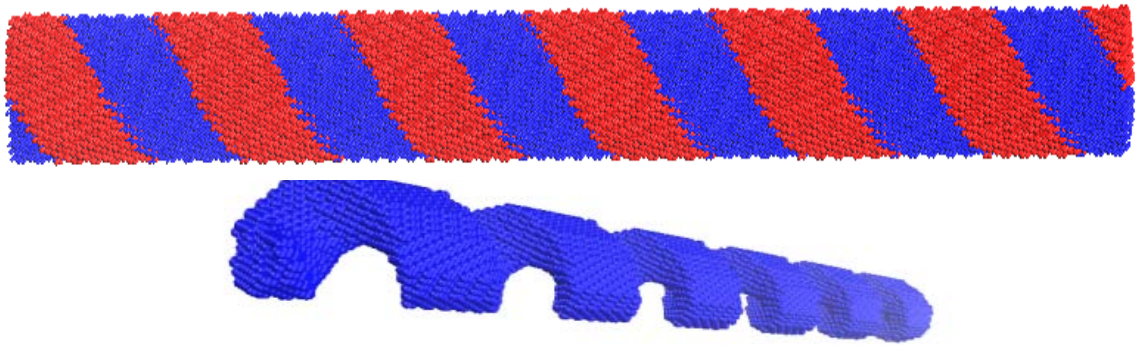

**Figure S1: *Top***: *Helical Structure of Type A (Si) and Type B (Si_4) systems for 6 helices of each type.* ***Bottom***: *Perspective view of same helical structure where only Type A atoms are shown.*

Representative graphs are shown below for the case with $n$ = 6 helices (*please refer to main text for 'n'*) and helical pitch length, $p$ ~ 4.6 nm, subjected to following series of relaxation simulations.

1. NVE simulation for 15 ns with velocity initialization at 600 K at the beginning of the simulation. Some of the kinetic energy is transferred to potential energy; hence the temperature does not reach 600K (as seen from Temperature graph below).
2. NPT simulation for 1ns (for rescaling temperature from 340 K to 300 K) followed by 5 ns of further NPT simulation to equilibrate the length of the nanowire at 1 atm.
3. Last NVE simulation for 3 ns to make sure the energy is still constant and temperature does not drift (as it was drifting in first NVE simulation).

As seen from the graphs below (*especially top left, for temperature*), we observed that in first NVE simulation, the temperature takes significant amount of time to reach constant value. The increase in temperature is complemented by decrease in potential energy (*top right*) to keep the total energy constant (*bottom left*). To make sure that temperature does not drift during later stages of the simulation (*which happens very often if the system of interest is not properly equilibrated*), we ran extended equilibration of 15 ns to avoid this issue. The rest of the simulations were performed to ramp the temperature further back to 300 K (*using NPT*) and to make sure that temperature do not drift in later stages of equilibration (*using NVE*) .

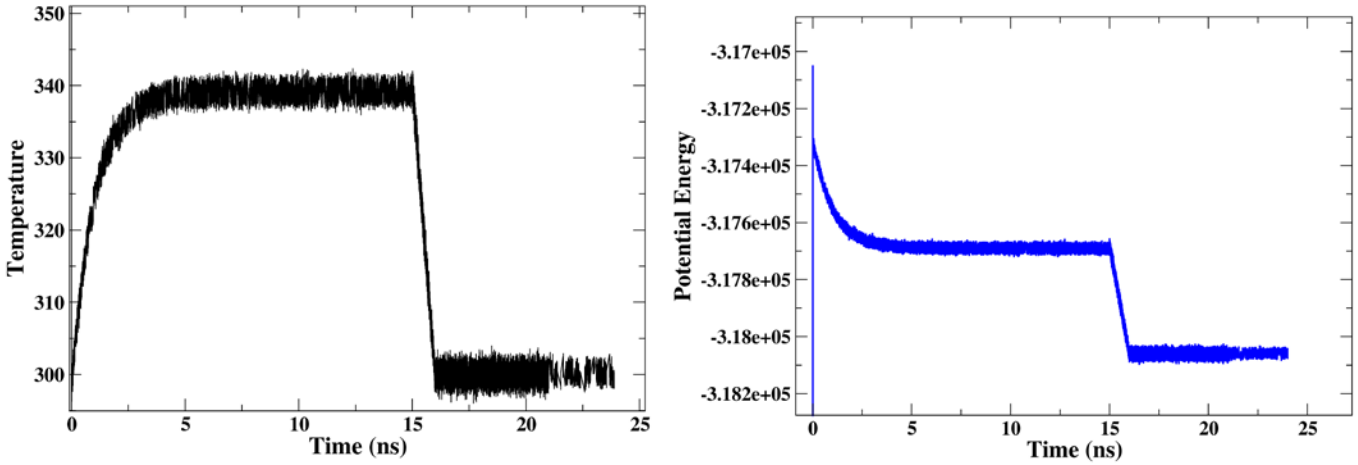


**Figure S2:** ***Left****: Temperature evalution of the nanowire as the function of equilibration time through different series of simulations.* ***Right****: Potential energy evaluation of the nanowire with respect to equilibration time.*

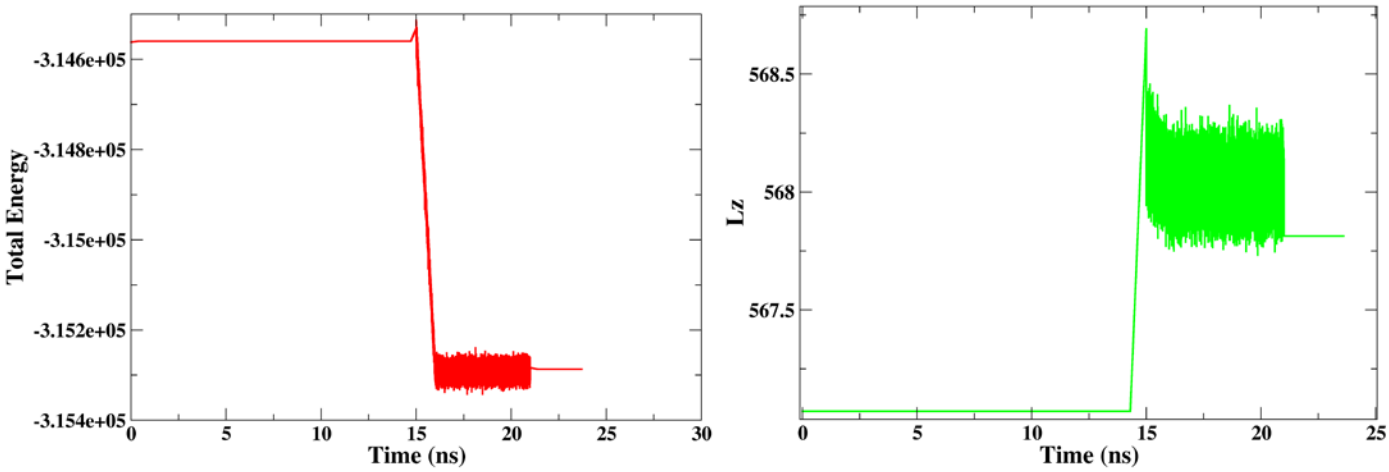


**Figure S3:** ***Left****: Total energy evalution of the nanowire as the function of equilibration time.* ***Right****: Evaluation of the length of the nanowire with respect to equilibration time.*

We believe that the necessity for prolonged simulations to achieve relaxation arises from ; a) large number of atoms in the system, and b) to attain full relaxation of surface atoms of the nanowire (as the initial nanowire was curved out from the bulk crystal).

## Supplementary Information B:

## Different Approaches to Simulate Thermal Transport using NEMD framework.

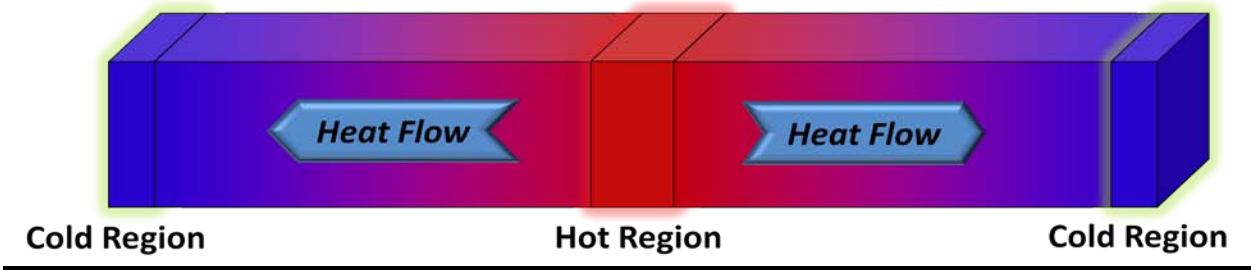


**Figure S4***: Schematic of non-equilibrium molecular dynamics methodology to calculate thermal conductivity*

### A. Approaches based on temperature thermostating of hot and cold regions (NVT schemes).

In all temperature controlled methodologies, the temperature of the hot and cold thermostats can be maintained using many different algorithms. In order to do so, certain amount of energy is provided (or taken off) to the thermostats which results in the heat flux (Q) across hot and cold thermostats. By maintaining the two thermostats at different temperatures, a spatial temperature profile is developed over time between the hot and cold regions. When the steady-state is reached, the ratio of heat flux (Q) with respect to temperature gradient provides the measure of thermal conductivity of the system. Below, we describe, in brief, the approaches that are commonly used to maintain the temperature of hot and cold thermostats.

**Method:** ***Temperature Rescaling (TR) (also known as velocity rescaling or isokinetic approach)***

**Brief description:** In this approach, the temperature of the hot and cold thermostats is controlled by rescaling the velocities of atoms that are present within respective thermostats, every *N* timesteps. Here, the rescaling factor is calculated by the ratio of current and target temperature. In other words,

$$v_{new} = v_{old}\sqrt{\frac{T_{new}}{T_{old}}}$$

It should be mentioned that the practice of simplistic rescaling of velocity, in general, perturbs the Maxwell velocity distribution in thermostated regions and samples incorrect microstates of NVT ensemble. So, frequency (N) of the rescaling is an important factor to consider while adopting this approach to make sure the velocity distribution is Maxwell-like in the thermostated region

**Variables:** Time interval (N) between temperature rescaling (Number of MD steps per velocity modification due to rescaling criterion)

**Method:** ***NVT Thermostating (using Berendsen Thermostat)[1]***

**Brief description:** In this approach, the temperature of hot and cold thermostat regions is controlled using Berendsen thermostat. This approach is a gentler version of *temperature rescaling (TR)* approach mentioned above. However, unlike TR, where the temperature is changed abruptly every N steps from $T_{old}$ to $T_{new.}$ Here, the temperature is gradually ramped towards desired temperature over N steps.

**Variables:** As in previous case, frequency (N) of temperature rescaling is something which one should consider to make sure results are not affected by it.

**Method:** ***NVT Thermostat (using Nose-Hoover Thermostat)***[2]

**Brief description:** While Berensen method or the velocity rescaling method is a convenient way to initialize the system temperature, it becomes important, once the system reached the desired temperature, that the system probes the correct canonical ensemble of microstates. Nose-Hoover method accomplishes this goal as an extended Lagrangian method, where the original system is treated as a part of an extended heat bath. The equations of motion in the original system are coupled with the rest of the extended system, and sample a canonical ensemble in the original system.

**Variables:** The damping coefficient, used in Nose-Hoover algorithm to modulate the temperature fluctuations is an important parameter to monitor. Often, a value of 100 (with respect to basic timestep of ~1fs) is used in this thermostat for temperature equilibration.

***Issue with approach A methodologies*:** In these approaches, the energy and momentum of the system are not conserved, in principle. While, on average, energy should be constant and overall momentum should be zero, this non-conservation can sometimes become an issue if proper care is not taken in defining the parameters (Often, this is the case when a large timestep is used). By using a smaller timestep, (~0.5 fs), the drift issues in energy and momentum can be taken care of in almost all cases.

## B. Approaches in which imposed heat flux is regulated (NVE Schemes)

In these approaches, the system remains in micro-canonical ensemble (NVE). However, the certain *a priori* known amount of energy (heat flux, Q) is transferred from one part (*cold region*) to another part (*hot region*) of the system. This energy transfer leads to the development of a temperature gradient between these regions. Once the steady-state is reached, the thermal conductivity can be predicted by the ratio of heat flux and the temperature gradient. Below, I briefly discuss two approaches that are often used within NEMD framework in NVE ensemble.

**Method:** ***Constant Heat Input/Output***

**Brief description:** This approach is very similar to temperature rescaling. However, the primary difference between two approaches lies in the way new target velocities are determined. In this approach, new target velocities are proportional to the ratio of square root of target energy to current energy, In other words,

$$v_{new} = v_{old}\sqrt{\frac{E_{old} \pm \Delta E}{E_{old}}}$$

where, ΔE is the energy transferred from cold to hot region.

**Variables:** The variables here are the heat flux Q, and the frequency (N) at which the kinetic energy ΔE is transferred between two regions.

**Method:** ***Muller-Plathe Algorithm (also known as Reverse-NEMD)[3]***

**Brief description:** When used appropriately, this is the only algorithm which conserved the energy as well as overall momentum of the system during the non-equilibrium molecular dynamics simulations. In this algorithm, the velocities of $N_{atoms}$ hot and cold atoms of 'same atomic mass' are switched between cold and hot thermostats respectively, every $N$ steps. As atomic masses are same, the velocity exchange does not alter the momentum of the system.

**Variables:** In original algorithm, the parameters which eventually determine the heat flux are the frequency (N) of velocity exchange between atoms of same mass and the number of atoms ($N_{atoms}$) involved in velocity swap at certain timestep. . However, recently, energy exchange between atoms of different masses has also been achieved with both energy and momentum conservation. This updated algorithm has recently been coded in LAMMPS MD simulation package.

***Issues with approach B methodologies*:** Although these approaches conserve energies and sometimes instantaneous momentum (Reverse-NEMD), one important concern associated with these methodologies is that the temperature of the hot and cold regions is not known a *priori.*

For example, in certain cases, the temperature of the hot and cold regions could be significantly different (~upto few hundred K) for low conductive materials if the heat flux is not properly tracked. In such cases, thermal conductivity values becomes little ambiguous when reported for specific temperature T.

Similarly, for comparative studies for different type of structures (like superlattice structure discussed in this study), a given set of $N$ and $N_{atoms}$ variables will result in significantly different temperature bounds (because of different thermal conductivity of different superlattice structures). Hence, comparing thermal conductivity values for such comparative studies at a well defined temperature become a little uncertain.

## Reason for choosing temperature rescaling methodology

The purpose of thermostat is to establish an external thermal stress. Our interest is in the response of the device between thermostats to the applied stress. Because the device itself is always in non-equilibrium state, the response of it to the applied stress is less sensitive to the integrity of the equilibrium distribution within the thermostats. Since, the results of thermal conductivity were not appreciably different when estimated by different methodology, the choice for picking one of the discussed techniques was convenience. First, we wanted to have a same temperature bound between the thermostats in our studies (which excluded NVE schemes). Among multiple NVT schemes, we choose the temperature rescaling methodology as we have had success with this methodology in our previous studies. A value of N=1000 (corresponding to 500 fs) between temperature rescaling was chosen to allow enough time for redistributing the updated velocities in Maxwell distribution.

1. Berendsen, H. J. C.; Postma, J. P. M.; van Gunsteren, W. F.; DiNola, A.; Haak, J. R., *J. Chem Phys*, **1984**, *81*, 3684.
2. Hoover, W. G. *Phys. Rev. A*, **1985**, *31* (3), 1695.
3. Muller-Plathe, F. *J Chem Phys*, **1997**, *106*, 6082.

## Supplementary Information C:

### Variance in Predicted Thermal Conductivity Values using Different Thermostat Methodologies

Below, we show the results for a sample study of bi-component helical nanowire corresponding to $n$ = 20 and how different thermostats affect the predicted value of thermal conductivity. As seen below, the maximum deviation that we observed was ~ 7%. Hence, we believe that for our systems of interest, thermostats did not affect thermal conductivity prediction significantly.

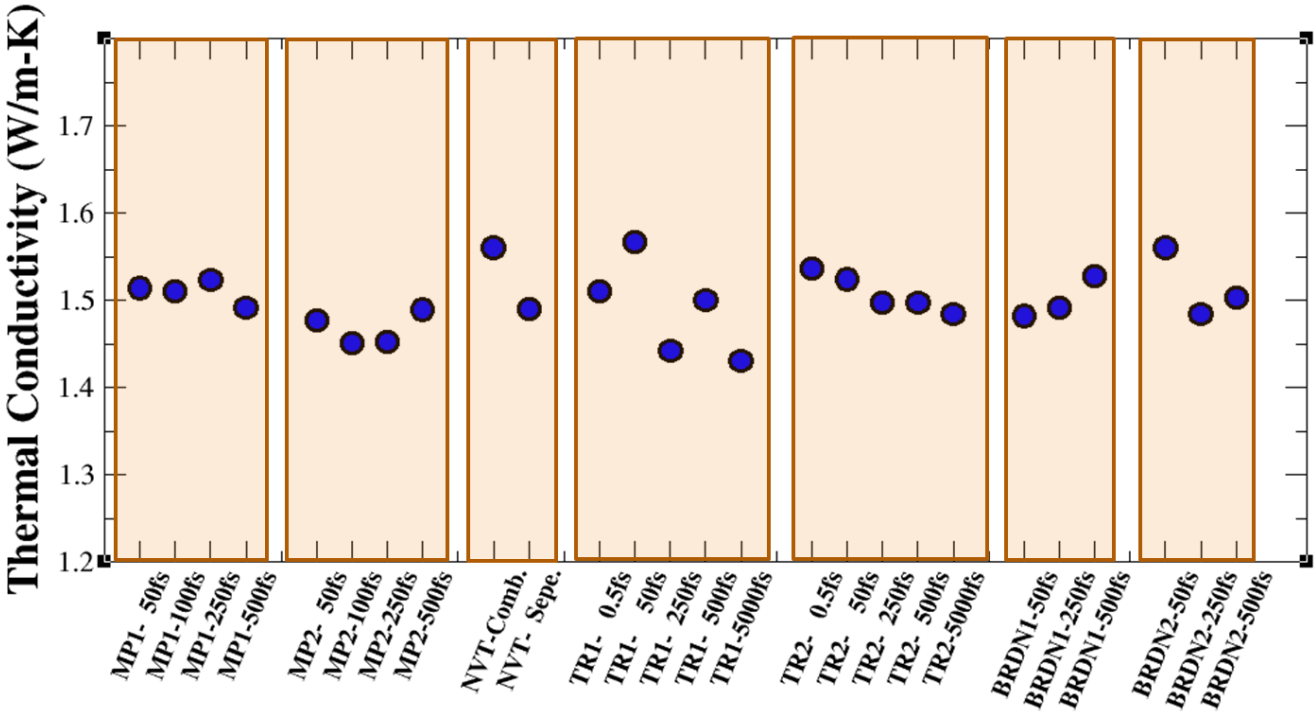


**Figure S5**: Comparison of thermal conductivity values as calculated using multiple NEMD approaches. The light shaded boxes are shown to separately highlight different methodologies employed in the simulations. The abbreviations in the abscissa are as follows: MP1(2):- Muller-Plathe algorithm in which velocity exchange occurs in only Si (both Si and Si_4); NVT-Comb (Sepe):- Nose-Hoover thermostating of the hot and cold regions. Comb (Sepe) corresponds to combined (separate) thermostats for two atom types; TR1(2):- Temperature rescaling approach for thermostating hot and cold regions. 1(2) corresponds to combined (separate) NVT thermostats for two atom types; BRDN1(2):- Berendsen thermostating of the hot and cold regions. 1(2) corresponds to combined (separate) thermostats for two atom types; XXXfs:- XXX corresponds to time interval for temperature rescaling or velocity exchange. Please refer to supplementary material B for detailed discussion.

## Supplementary Information D:

## Effect of Mass Ratio on the Thermal Conductivity of Helical and Superlattice Nanowires: Detailed Plots

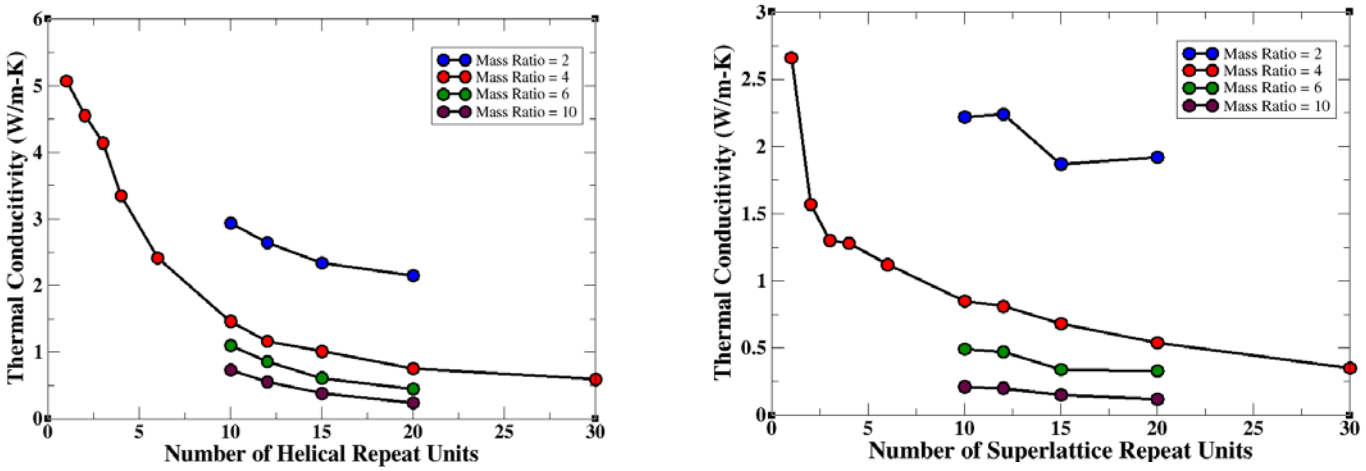


**Figure S6:** Thermal Conductivity of (a) Helical and (b) Superlattice nanowires with respect to number of repeat units

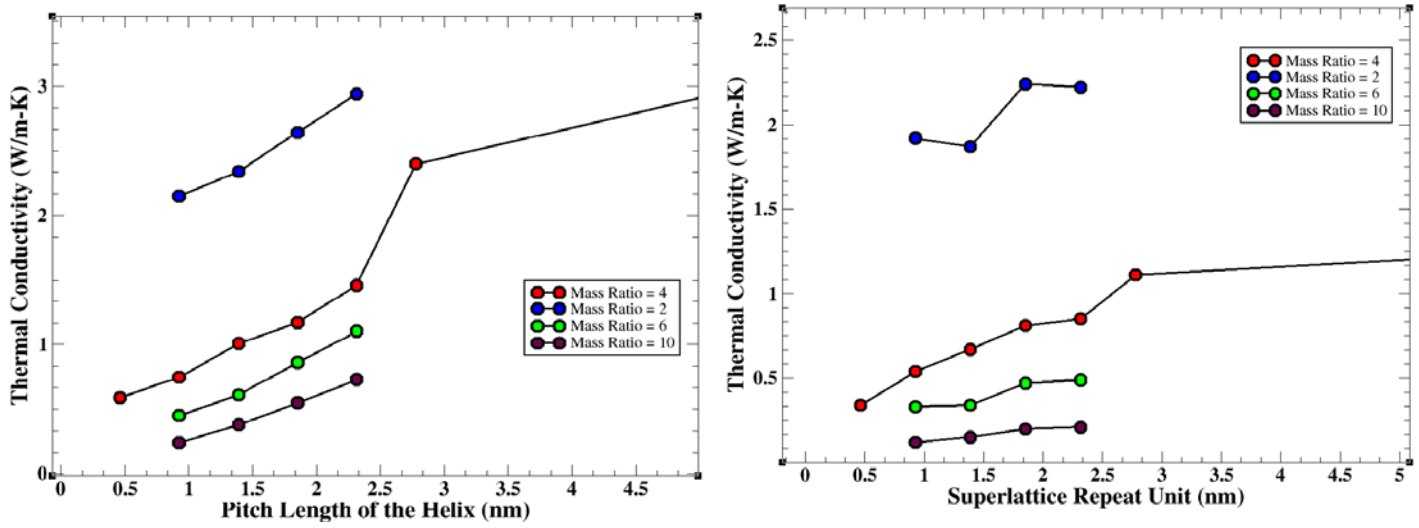


**Figure S7:** Thermal Conductivity of (a) Helical and (b) Superlattice nanowires as a function of pitch length or period of the superlattice.