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Intrinsic Localized Modes in Strained Graphene Sheet

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Abstract—Dynamics and stability of intrinsic localized modes (ILM) in graphene sheets are investigated. Structure of ILM is investigated by numerical solution obtained by a method coupling molecular dynamic (MD) method and iteration method. Moreover linear stability of ILM in the graphene sheet under strain is presented, based on the Floquet theory.

1. Introduction

Intrinsic localized mode (ILM) or discrete breather (DB) has extensively attracted in nonlinear physics recently[1, 2]. ILM is a temporal-periodic, space-localized structure which is excited in nonlinear lattices. Since ILM is vibration due to nonlinearity of systems, vibrational frequency is out of linear phonon bands which are produced due to discreteness of the systems. Various studies[3] have been done not only in theoretical aspects such as existence, stability and mobility, but also applications in real systems such as large structures[4], mechanical systems[5] and micromechanical systems[6].

It is well known that structure of atoms in crystalline can be modeled as a lattice model. Phonon band, which is produced from discreteness of the lattice model, contains important properties of solid states of crystals. Interaction potential which determines dynamics of crystals is intrinsically anharmonic. Nonlinear dynamics can appear when displacement of atoms becomes large. Therefore ILM can be excited in crystalline.

Excitation of ILM in materials has been studied in various aspects. Marín has investigated numerically excitation of moving ILMs in a 2D hexagonal lattice with Lenard-Jones potential and on-site potential, which is a model of layered materials[7]. Cuevas has studied interaction of ILM and impurities such as vacancies and interstitial defects in 1D Frenkel-Kontorova (FK) lattices[8]. They have also investigated migration of vacancies due to collisions between vacancies and ILMs[9].

Molecular dynamics (MD) method is widely used for investigating mechanical properties in atomic scale of material. Since interaction potential which is heuristically given has nonlinearity, Excitation of ILM can be observed in MD simulations. Yamayose[11] has studied excitation of ILM in graphene sheets, which is 2D structure of carbon atoms. ILM excitation in carbon nanotubes (CNTs) with specific chirarities has been also reported[12, 13]. Shimada has reported that ILM plays an important role in nucleation of defect's in CNTs such as Stone-Wales transformation[14].

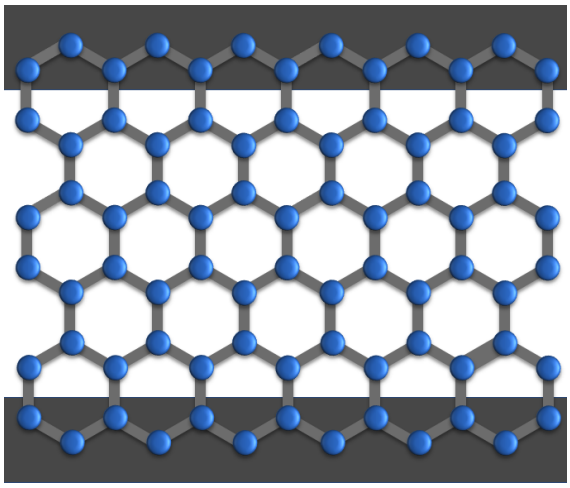

Figure 1: Schematic description of model

It is well known that mechanical properties of materials can be usually described as a response of stress and external force acting of the materials. In microscopic view, arrangement of atoms can change. ILM can also be affected by this rearrangement of atoms, i.e. strain of lattices.

ILMs excited in MD simulations have finite lifetime. It should be noted that ILMs excited in MD simulation is not a exact numerical solutions, since they are perturbed by thermal fluctuation of surrounding atoms. Constructing the exact numerical solution of ILM is useful for investigating basic properties of ILM. For example, once we obtain the numerical solution, it is possible to study their stability.

In this study, we propose the numerical method for obtaining the exact numerical solutions in crystals by coupling MD method and iteration method. We also investigate the linear stability of numerical solutions of ILMs by Floquet theory of periodic solutions. This method is applied to the graphene sheet which is strained by the external forces.

2. Model

We consider two dimensional hexagonal lattice which are consist of carbon atoms. Atom's motion is limited in the plane. Fig. 1 shows an example of models. Periodic bound-

ary condition is considered in x-direction. Top and bottom boundaries are fixed. Fixed boundaries are displaced in order to give strain to the system. Thermal bath is not connected to the system.

Interaction of atoms is modeled by a interaction potential proposed by Brenner[15]. Hamiltonian of Brenner potential is written as follows:

$$H = \sum_{i}^{N} \sum_{\alpha} \frac{(p_i^{\alpha})^2}{2m} + \frac{1}{2} \sum_{i}^{N} \sum_{j \neq i}^{N} \sum_{k \neq i,j}^{N} \Phi_{ijk}(r_{ij}, r_{ij}, \theta_{ijk}), \quad (1)$$

where m is mass of the carbon atom, N is number of atoms, i, j, and k are the indices of the carbon atoms, α indicates the coordinates x and y, p_i^{α} is momentum of the atoms, Φ_{ijk} is a interatomic potential, $r_{ij} = x_j - x_i$, $r_{ik} = x_k - x_i$ is a distance between atoms i-j, and i-k, θ_{ijk} is the angle between two bonds i-j and i-k.

3. Numerical Method

3.1. Searching Numerical Solution

Displacement and momentum of N atoms in a graphene sheet can be described $4N$ variables. Let $\mathbf{X} = \{x_i, p_i\}$ be state variables of the graphene sheet. Temporal evolution of the state variables can be described by the equation of motion

$$\dot{\mathbf{X}} = f(\mathbf{X}; H), \quad (2)$$

where f is a function of $\mathbf{X}$ and depends on H. Let $\mathbf{A}(t_1; \mathbf{X}(t_0))$ is a map of the state variables from time t_0 to $t_0 + t_1$:

$$\mathbf{X}(t_0 + t_1) = \mathbf{A}(t_1; \mathbf{X}(t_0))\mathbf{X}(t_0). \quad (3)$$

ILM is a time-periodic solution. Therefore ILM takes a periodic orbit in the phase space. Let T be a period of ILM. A state variable $\mathbf{X}_{\mathrm{ILM}}$ which is on the periodic orbit satisfies a relation $\mathbf{X}_{\mathrm{ILM}}(t + T) = \mathbf{X}_{\mathrm{ILM}}(t)$. Therefore we obtain an equation:

$$\mathbf{X}_{\mathrm{ILM}}(t) = \mathbf{A}(T; \mathbf{X}_{\mathrm{ILM}}(t))\mathbf{X}_{\mathrm{ILM}}(t), \quad (4)$$

for any t.

Searching a numerical ILM solution with a period T is equivalent to solving equation (4) for T. We search the solution of (4) by the conjugate gradient method. The map $A(T; \mathbf{X}_{\mathrm{ILM}}(t))$ can be obtained by the temporal evolution of equation (2). This temporal evolution is done by MD simulation of the graphene sheet. We search ILM solution following procedure:

1. Determine a initial guess of a ILM solution of equation (4) with period T.
2. Calculate the temporal evolution during T from the guess by performing MD simulation.
3. Evaluate the difference between the guess and the result of temporal evolution.
4. If the difference is smaller than tolerance, we determine the guess is the solution. If not, new guess is calculated by conjugate graduate method and we return to 2.

Equation (2) has $4N$ unknown variables. It is important to make a good initial guess for obtaining convergence solutions. Therefore we firstly search the solution of the system with small degree of freedom. Then we increase the degree of freedom and use the previous solution as the initial guess of new system.

3.2. Stability Analysis

Once we obtain the ILM solution $\mathbf{X}_{\mathrm{ILM}}$, we can analyse linear stability of the ILM. Let ξ be a small perturbation between $\mathbf{X}_{\mathrm{ILM}}$. The variational equations for ξ is given by

$$\dot{\xi} = \left. \frac{\partial f(\mathbf{X}; H)}{\partial \mathbf{X}} \right|_{\mathbf{X}=\mathbf{X}_{\mathrm{ILM}}} \xi. \quad (5)$$

Coefficients of equation (5) are T-periodic. Stability of the perturbation ξ can be determined by checking properties of the monodromy matrix $\mathbf{M}$. The monodromy matrix is defined as

$$\xi(T) = \mathbf{M}(T)\xi(0). \quad (6)$$

If the monodromy matrix $\mathbf{M}$ has an eigenvalue larger than unity, the perturbation becomes unstable towards the corresponding eigenmode.

Monodromy matrix $\mathbf{M}$ cannot be obtained explicitly. Instead, we can calculate $\mathbf{M}$ numerically. Consider 4N vectors $\xi^{(i)}(0)$ whose components are zero except that i-th component is unity. $\mathbf{M}$ is given by the

$$\mathbf{M} = (\xi^{(1)}(T), \xi^{(2)}(T), \cdots, \xi^{(4N)}(T)), \quad (7)$$

where $\xi^{(i)}(T)$ is the temporal evolution from $t = 0$ to $t = T$ of the i-th vector.

Note that we have to calculate the ILM solution $\mathbf{X}_{\mathrm{ILM}}$ in integrating the variational equations (5).

4. Results and Discussion

Fig. 2 shows structure of ILM obtained by the iteration method. Color of atoms indicates the kinetic energy of the atoms (blue is low and red is high). Two carbon atoms which are connected by a bond vibrate along the direction parallel to the bond. Surrounding atoms vibrate slightly. Period of the vibration is 19ps. This structure of ILM is the same as those reported in [11]. This type of ILM seems to be one of the most typical structures of ILM excited in the graphene sheet.

Fig. 3 shows the temporal evolution of the displacement of the two atoms by MD method. The numerical solution of ILM is used as a initial condition. In this figure,

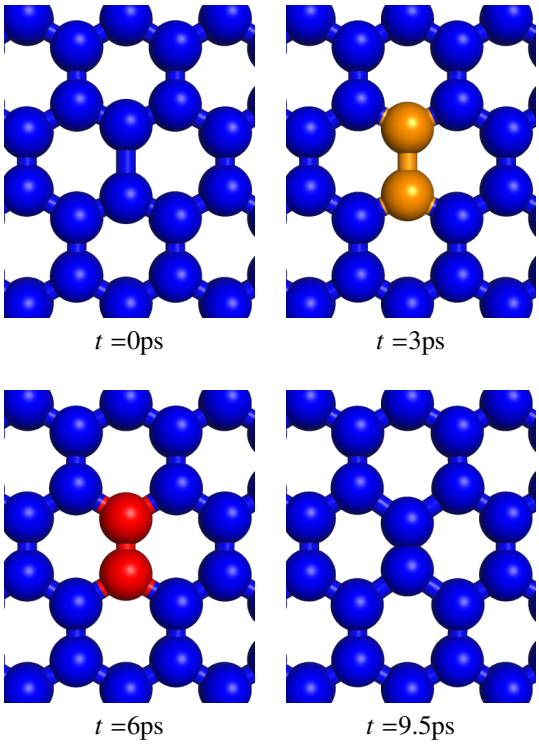

Figure 2: Snapshot of structure of ILM. Color indicates temperature of the atom.

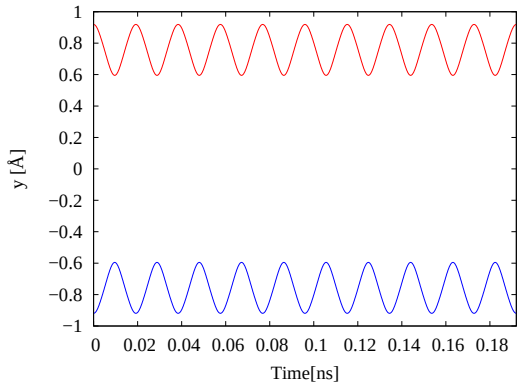

Figure 3: Displacement in y-direction of two atoms in ILM

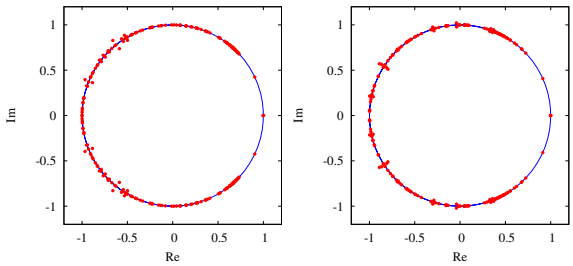

Figure 4: Distribution of eigenvalues of ILMs: left no strain, right 10% strain

we show dynamics of atoms during about 0.2ns. However, we observe that the numerical solution of ILM alive about 10ns duration. Angular frequency of ILMs is larger than the maximum angular frequency of the phonon band[11]. Amplitude of the vibration of bond is about 0.3Å, which is about 20% of the equilibrium length of the bond.

We calculate the ILM solution in graphene sheets applying the strain in y-directions. We introduce strain to the graphene sheet from 0% to 10%. At first, the system is relaxed by minimizing the total energy by using conjugate graduate method. Nest, we search the ILM solution from initial guess which two neighboring atoms are displaced with large amplitude.

We find that structure of ILM does not change drastically. Amplitude of ILM of the same period becomes larger when the strain of the system becomes larger. Amplitude of ILM grows mainly in stretching direction. Growth in compressing direction is small, since larger gradient of the interaction potential between atoms against compression than that against stretch.

Effect of strain mainly appears in stability of ILMs. Fig.4 shows the distribution of eigenvalues of monodromy matrix in the complex plane in the cases of no strain and 10% strain. It is found that distribution of eigenvalues varies as the strain is introduced. Some unstable perturbation modes also vary its values. Therefore the unstable mode which has largest growth rate can be also varies. This fact leads to change of structure of the most unstable modes.

We can classify the most unstable mode into three cases in the view of their pattern. Fig. 5 shows displacement pattern of three cases. In the case that strain is less than 2%, atoms surrounding ILM become unstable but atoms in ILM do not. In the case that strain is from 2% to 5%, atoms in ILMs can also become unstable in one mode. In the case that strain is greater than 6%, atoms in ILM are unstable in both modes.

It is interesting that this unstable mode produce shear motion near ILM. It can be seen that a pair of three atoms in near ILM move in opposite direction. In [14], cutting and rebonding between carbon atoms due to ILM is reported. Excitation of unstable mode which has shear motion can be an explanation for a mechanism of cutting and rebondig phenomena.

5. Conclusion

In the present study, we proposed the numerical method for obtaining solution of ILM in crystals by coupling MD method and irrational method. We also show the stability analysis of ILM in crystals. Numerical solutions obtained by the proposed method have good accuracy for dynamic simulations. We investigate the structure and stability of ILM in the strained graphene sheet. It is found that the unstable mode excited in the strained case leads to shear motion near atoms near ILMs.

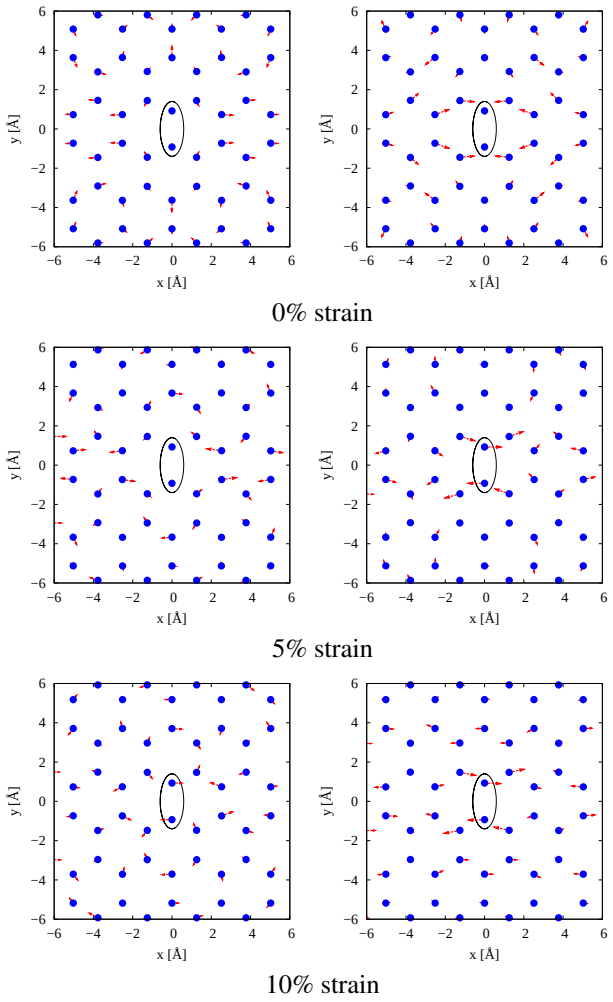

Figure 5: Unstable perturbation mode with the maximum growth rate. Atoms inside black circle are in ILM

Acknowledgments

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