Intrinsic localized modes and trapped phonons in crystal lattices

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Abstract. An analytical theory of intrinsic local modes (ILMs) in infinite perfect lattices is proposed. One prediction of the theory is that for some conditions linear local modes appear near the ILM. High-precision MD calculations of the vibrations of diatomic chains confirm the prediction.

It is already a well-known fact that localized vibrational excitations can exist in perfect anharmonic lattices [1-10]. Such excitations are called intrinsic localized modes (ILMs) [1], discrete breathers [6] or discrete solitons [10]. The main approach, which has been used for the study of these vibrational excitations, is molecular dynamics (MD) simulations, which is rather efficient in the case of simple 1D lattices, but requires very lengthy computations in the case of 2D and 3D lattices. Therefore, it is of interest to develop other methods which would allow one, when studying ILMs, to reduce the amount of numerical computations.

It is to be expected that the appearance of an ILM would change the local properties of the lattice including the local phonon dynamics. This back reaction on the phonon spectrum should have physical consequences since an ILM could induce linear local modes (LLMs) outside the plane wave spectrum (see Reference [11] where the existence of the LLMs trapped by an ILM have been demonstrated for a monatomic chain with hard quartic anharmonicity).

The goal of this communication is to develop a theory of ILMs and LLMs, for macroscopically large lattices of arbitrary dimensions and realistic interaction potentials. We start with the classical equations of the motion

\[ M_n \ddot{u}_n = -\partial V / \partial u_n, \]

where \( u_n \) is the displacement of the atom \( n \), \( M_n \) is its mass (the subscript \( n \) includes both the site number and the number of the Cartesian component), \( V \equiv V(\{u_n\}) \) is the potential energy:

\[ V = \sum_{\text{nn}} V^{(2)}_{\text{nn}} u_n u_n / 2 + V_{\text{anh}}, \]

where the first term is its harmonic part and the second term is its anharmonic part. We are considering a localized solution of the equations of motion, describing a stable ILM. For such a solution the displacements of atoms are of the form

\[ u_n(t) = A_n \cos \omega_n t + \xi_n + O(\omega_n), \]

where \(|A_n|\) is the amplitude of the ILM at the site \( n \).

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\( \omega_L \) is the frequency of an ILM, which lies outside the phonon spectrum, \( \xi_n \) is the shift of the equilibrium position of the atom \( n \) (i.e. the dc-component of the ILM; it differs from zero due to odd potential terms), \( O(\omega_L) \) is the sum of higher harmonics. The contribution of these harmonics is small if the amplitude of the ILM is small as compared to the lattice constant(s). Below these terms are neglected. Note that \( \tilde{u}_n = A_n \cos(\omega_L t + \xi_n) \) is also a solution of the equations of motion. Taking \( \tau \) to be infinitesimal, we get \( \tilde{u}_n(t) = u_n(t) + q_n(t) \), where \( q_n = -A_n \omega_L \tau \sin \omega_L t \). Subtracting the equation for \( u_n \) from that for \( \tilde{u}_n \), one gets the equation for \( q_n(t) \)

\[
M_n \ddot{q}_n = -\sum_n (V^{(2)}_{nn} + v_{n,m}q_m)q_n, \tag{1}
\]

where \( v_{n,m} = \partial^2 V_{nm}/\partial u_n \partial u_m \). We are searching for the solution of the equation (1), which has the \( \sin \omega_L t \)-type time dependence. Let us apply the rotating wave approximation (RWA) and extract the \( \propto \sin \omega_L t \) term in the product \( v_{n,m}q_m \). Doing that, we find the following equation for the time-independent part of \( v_{n,m} \): \( v_{n,m} = 2 \left\{ v_{n,m} \sin^2 \omega_L t \right\} \), where \( \left\{ \ldots \right\} \) denotes the averaging over time. The perturbation \( v_{n,m} \) should be determined self-consistently. This can be done by using the Lifshitz method [12]. In this method, the ratios of the amplitudes of a local mode (in our case these are equal to the ratios of the amplitudes of the ILM) satisfy the relations

\[
A_n/A_0 = G_{n0}(\omega_L)/G_{00}(\omega_L), \quad G(\omega) = \left( I - G^{(0)}(\omega) \nu \right)^{-1} G^{(0)}(\omega), \tag{2}
\]

(\( G(\omega) \) is the perturbed Green’s function), the frequency \( \omega_L \) is given by the position of the pole of \( G(\omega) \) outside the phonon spectrum determined by the equation \( I - G^{(0)}(\omega_L) \nu = 0 \). Since the amplitude parameters \( A_n \) enter into the perturbation matrix \( \nu \), the equations (2) constitute self-consistency equations.

The dc-shifts \( \xi_n \), entering into Eqs. (2), are not independent parameters of the problem; they can be expressed through \( A_n \). The relation between \( A_n \) and \( \xi_n \) reads \( \xi_n = \sum'_n g_{nm} \left\{ \partial V_{nm}/\partial u_m \right\} \), where \( g_{nm} = G_{nm}^{(0)}(0)/\sqrt{M_n M_m} \) are the parameters of the harmonic lattice, \( G_{nm}^{(0)}(0) \) is the static limit (\( \omega = 0 \)) of the Green’s function of the perfect lattice.

To calculate an ILM, we start by choosing its main amplitude(s). By using these, we find the dc-shifts \( \xi_n \). By inserting these \( \xi_n \) into Eq. (2), we calculate the matrix \( \nu \). After that we compute \( \omega_L \) and then the new amplitudes. We repeat the procedure with newly-found amplitudes until the amplitudes converge to the required accuracy. Our examples show that the procedure usually converges rather quickly. After the ILM is calculated, one can find the LLMs. In the RWA the latter modes are also described by Eq. (1), where instead of \( \sin \omega_L t \) one has \( \sin \omega_L t \); \( \omega_L \) is the LLM frequency. Due to the difference of \( \omega_L \) and \( \omega_L \) the time-averaging can be performed independently for the ILM and for the LLM. As a result, instead of \( v_{n,m} \) one gets \( v_{n,m} = \left\{ \partial^2 V_{nm}/\partial u_n \partial u_m \right\} + \lambda A_n A_m \).

Here the \( \propto \lambda \) term takes into account the orthogonality condition of the LLM with the ILM. If the ILM and the LLM have different symmetry, then \( \propto \lambda \)-term is absent.

Let us apply the presented theory to a diatomic chain with the Born-Mayer-Coulomb pair-potential:

\[
V(x) = \left( q^2 / d^2 \right) \left[ \rho e^{-x/p} - \left( d^2 / (d + x) \right) \right] + d - \rho.
\]

Here \( q \) is the effective charge, \( \rho \) gives the strength of the repulsion of atoms, and \( d \) is the
lattice constant. It is known [6] that in this model there exist odd ILMs centred on light ions.

For numerical considerations we choose the potential parameters of a SrO-like chain, and calculate the odd ILMs centred on a O atom. The parameters of this potential are known: \( d = 2.58 \text{Å} \), \( \rho = 0.37 \text{Å} \), \( q = 2e \). In this case, the top of the acoustic band is at \( \omega_a = 0.412 \omega_m \), the bottom of the optical band is at \( \omega_o = 0.964 \omega_m \); \( \omega_m \) is the highest phonon frequency. We take into account the nearest and the next-to-nearest interactions. In this chain, together with an odd ILM, mostly localized on the central oxygen ion, with the frequency in the gap of the phonon spectrum, one gets the even pocket-mode-type LLM with the frequency \( \omega_l \) slightly above the phonon spectrum, localized mostly on next neighbor oxygen atoms. Note the different directions of the shifts of the ILM and the LLM frequencies with respect to the optical phonon band. This results from the difference of the perturbation \( v' \) which leads to the LLM from the self-consistent perturbation \( v \) which determines the ILM. The theoretical values of the ILM frequency for \( A_0 = 0.7 \text{Å} \) and \( A_0 = 1.0 \text{Å} \) equal respectively \( 0.795 \omega_m \) and \( 0.710 \omega_m \). The theoretical values of the LLM frequency in these cases are \( 1.0008 \omega_m \) and \( 1.007 \omega_m \), respectively.

![Figure 1.](image)

**Figure 1.** The power spectra of ILMs+LLMs in the SrO diatomic chain with the amplitude parameter \( A_0 = 0.7 \text{Å} \) (left) and \( A_0 = 1.0 \text{Å} \) (right). The bottom panels correspond solely to ILM; the upper panels, to the spectra in the case of small additions of LLM to the ILM. Dotted lines are the borders of the phonon bands.

We have also performed molecular-dynamics (MD) simulations of ILMs and LLMs in the SrO-
like chain with $\sim 10^5$ atoms and calculated the power spectra of vibrations between 2700 $T$ and 3000 $T$ ($T$ is the period of the ILM). Examples of the calculated power spectra are given in the bottom panels of figure 1, where the spectra for two different amplitudes of the central atom are shown. Each spectrum has a strong single peak with the frequency $\omega_L$ in the gap of the phonon spectrum. The MD values of the ILM frequency for $A_0 = 0.7\text{Å}$ and $A_0 = 1.0\text{Å}$ are 0.799$\omega_m$ and 0.719$\omega_m$, in very good agreement with the theory. When small additional amplitude shifts, appropriate to the even pocket-mode-type LLM, are added to the ILM amplitude pattern, the additional small peaks in the power spectrum are obtained near the top of the phonon spectrum (see the upper panels in figure 1). The frequency as well as the relative amplitudes of this weak linear vibration are in very good agreement with the theory of LLMs presented here. The frequencies of two other even weaker peaks equal $\omega_L - \omega_L$ and $2\omega_L - \omega_L$; these peaks result from the nonlinear mixing of the ILM and LLM.

To sum up, we have presented a theory that allows one to describe ILMs and the effect of ILMs on phonons. The theory is applied to a SrO-like 1-D chain with Born-Mayer-Coulomb interaction potentials. Given sufficient amplitude the theory predicts the appearance of an ILM in the gap and of an LLM above the top of the plane wave spectrum. The theoretical findings are in good agreement with MD simulations.

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