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Modeling of self-localized vibrations and defect formation in solids

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ABSTRACT

MD simulations of recoil processes, following the scattering of X-rays or neutrons are performed. At small energies (<10 eV) the recoil can induce intrinsic localized modes and linear local modes associated with them. In ionic crystals the frequencies of these modes are located in the gaps of the phonon spectrum, being essentially dependent on long-range forces. In metals, as a result of the screening of atomic interactions by free electrons, their frequencies can be positioned above the phonon spectrum. The MD simulations of vibrations in Ni and Nb confirm this prediction. If the recoil energy exceeds tens of eV, the vacancies and interstitials can be formed. In fcc lattices a recoil in (1 1 0) direction can produce a vacancy and a crowdion, while in the case of a recoil in (1 0 0) and in (1 1 1) directions a bi-vacancy and a crowdion can be formed.

1. Introduction

The goal of the present communication is to study the motion of atoms (ions) in crystals, caused by the recoil processes following the scattering of X-rays or neutrons. We are considering the case of moderate recoil energy (0.1 eV < E_R < 200 eV). In this range of energy one can observe the creation of intrinsic localized modes (ILMs) [1–3] and the linear local modes (LLMs) associated with them [4], if E_R < 10 eV and the creation of defects (vacancies, interstitials, crowdions), if E_R > 10 eV. In the case of ionic crystals we suppose that in the given range of energies, the creation of electron–hole pairs and excitons in the scattering processes can be neglected.

In Refs. [5–7] we introduced a method of calculations of ILMs and used it for the MD simulations to study nonlinear dynamics of Ni and Nb. Below we report the results of the investigation and show that in these metals ILMs and the LLMs stemming from them can be positioned above the phonon spectrum. We also discuss the recoil-induced ILMs [8] and the radiation defects near the low-energy threshold in the alkali-halide crystals NaI and KCl. In these crystals the usual cluster methods of the MD simulations of ILMs may become unsatisfactory unless in the case when the size of the clusters is very large ($\sim 10^6$ atoms or more). Indeed, at the energies E_R < 10 eV (ILM region), the dynamics of atoms (ions) in ionic crystals is governed by both linear and non-linear forces. The latter forces are usually short-range ones, while the linear forces in non-metallic systems, as a rule, are essentially long-range (LR) ones. The situation differs from the impact-induced defect formation at E_R > 10 eV, when the acting forces are essentially short-range ones. We have developed a method which allows the LR forces to be taken into account. In this method, LR forces are included in the calculations via the phonon Green's functions for the infinite crystal, while the anharmonic forces of a localized excitation are considered explicitly within a finite cluster. As in 3D lattices, both the actual Green's functions and anharmonic forces are localized and the numerical solution of the equations of motion is possible.

2. ILMs and LLMs in 3D lattices

2.1. Self-consistent theory of ILMs and LLMs

An ILM corresponds to a solution $u_L(t) = A_n \cos(\omega_L t) + O$ (the term O describes small higher-order harmonics). The amplitudes A_n and the frequency ω_L can be found from a self-consistency consideration [5–7] solving the following linear equation for the small variations (α_n) of amplitudes:

$$\ddot{\alpha}_n = -\sum_{n'} (D_{nn'} + w_{nn'})\alpha_{n'} \quad (1)$$

Here $D_{nn'}$ is the dynamical matrix of the harmonic lattice, $w_{nn'}$ is the perturbation caused by the ILM. The solution is $\alpha_n/\alpha_0 = \tilde{G}_{n0}/\tilde{G}_{00}$, where

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$$\tilde{G}(\omega) = \left(I - G^{(0)}(\omega)w\right)^{-1} G(\omega) \quad (2)$$

$G(\omega)$ is the Green's function of phonons in a spectral representation.

A small variation of the ILM satisfies the condition $\alpha_n = \zeta \cdot A_n \sin \omega_L t$, where ζ is a small parameter (note the phase shift $\pi/2$ with respect to the ILM). However, this condition does not apply to other variations. Therefore the values of $w_{nn'}$ for the ILM and for the perturbed phonons are different. In Ref. [4], we have shown that the latter perturbation can lead to the localization, producing linear local modes (LLMs). These modes cause the modulation of ILMs (see Fig. 1). Equations for $w_{nn'}$ are

$$w_{nn'} = 2\langle \sin^2 \omega_L t \partial^2 V_{anh}/\partial u_n \partial u_{n'}\rangle, ILM \quad (3)$$

$$w_{nn'} = \langle \partial^2 V_{anh}/\partial u_n \partial u_{n'}\rangle + \lambda A_n A_{n'}, LLM, \quad (4)$$

where λ is the Lagrange multiplier which should be found from the orthogonality condition of the LLM under consideration. Eqs. (1)–(4) present a set of self-consistent equations, which can be solved numerically by using, e.g. the iteration procedure [4–7].

The presented theory was applied for the calculation of ILMs and LLMs in a monoatomic chain with quartic anharmonicity and for alkali halide crystals. The MD simulations of ILMs and LLMs were also performed for these systems [4–7]. The results of both considerations are in very good mutual agreement. Recently, the LLMs, predicted by us, have been experimentally observed in Ref. [9].

2.2. ILMs and LLMs above the phonon spectrum

In the numerical studies of ILMs, different pair potentials, such as Lennard–Jones, Born–Mayer–Coulomb, Toda, and Morse potentials, have been used. All these potentials show a strong softening with an increasing vibrational amplitude. The ILMs, which have been found in these simulations, always split down from the optical band(s) into the phonon gap, if there is one. (see Refs. [10,11], where ILMs in alkali halide crystals have been calculated). However, a recent inelastic neutron scattering investigation of the vibrational excitations in metallic uranium (α-U) showed some degree of localization near the top of the phonon spectrum at elevated temperatures [12]. For such a phenomenon to occur, the pair potentials in this metal must be very different from the ones describing alkali halide crystals. As the free electrons at the Fermi surface provide an essential contribution to the screening of the ion–ion interaction in metals, there is no *a priori* reason to expect the anharmonicities of these two very different systems to be similar.

We have applied the above-presented theory to study ILMs in metallic nickel and niobium. We have used the embedded atom model (EAM), allowing one to take the screening effects into account. Our findings show that ILMs and LLMs may be observed above the phonon spectrum if [7]

$$\bar{\kappa} = 3k_4\bar{k}_2/4k_3^2 > 1 \quad (5)$$

Here k_3 and k_4 are the cubic and quartic anharmonic springs, $\bar{k}_2 = Mv^2/r_0^2$ is the mean elastic spring in the bulk, v is the longitudinal velocity of sound, M is the mass of atoms, r_0 is the lattice constant. We have used the potentials given in [13,14] and have found that in Ni and Nb this condition is fulfilled. This allowed us to conclude that in both these metals one can observe ILMs with the frequencies above the phonon spectrum. This conclusion was verified by the MD simulations of ILMs in these crystals. In Fig. 1 the time dependence of the vibrations in metallic Ni at the recoil energy E_R = 0.5 eV is presented, while in Fig. 2 the dependence of the ILM frequency on the amplitude of the central bond is given. In Fig. 2 the main oscillations correspond to the ILM; the periodic modulation of the amplitude of the ILM is caused by the LLM. Frequencies of both, ILM and LLM exceed the top phonon frequency.

3. ILMs in 3D lattices, long-range forces included

The vibrational dynamics of dielectrics can usually be described in the adiabatic approximation with the potential energy of the lattice being the sum of all pair potentials. In this approximation, the displacement of the nth atom (ion) is described by the equation

$$u_n(t) = u_n^{(0)}(t) + \int_0^t d\tau \sum_{n'} G_{nn'}(t-\tau) F_{n'}^{anh}(\tau)(M_n M_{n'})^{-1/2}, \quad (6)$$

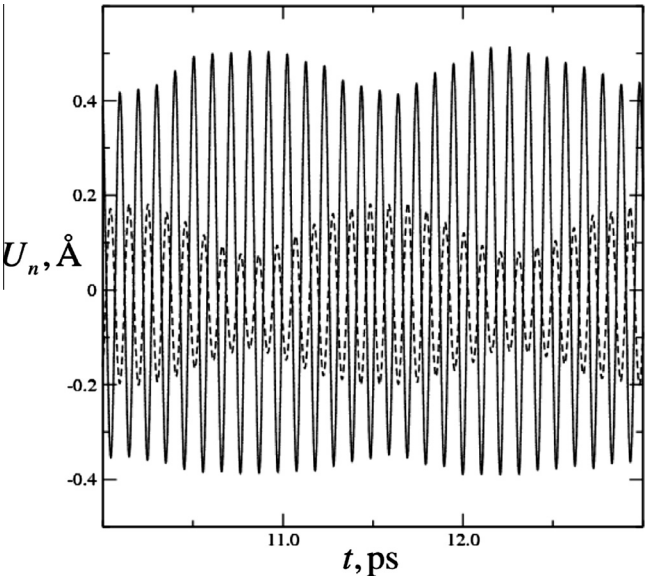

Fig. 1. Even ILM in Ni: oscillations of two atomic bonds: central (solid line) and the third one from the centre (dashed line). Modulation of amplitudes is caused by the LLM.

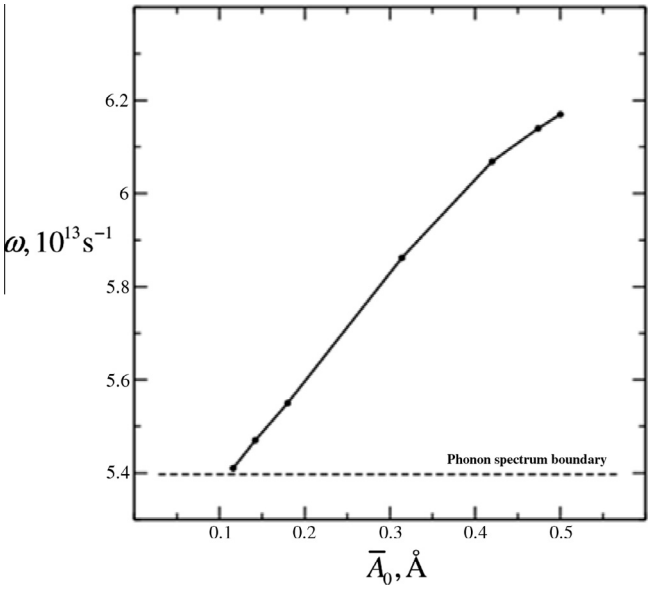

Fig. 2. The dependence of the frequency of the even ILM in Ni on the amplitude of vibrations of the central bond.