Linear local modes induced by intrinsic localized modes in a monatomic chain

V. Hizhnyakov and A. Shelkan*

Institute of Physics, University of Tartu, Riia 142, 51014 Tartu, Estonia

M. Klopov

Institute of Physics, Tallinn University of Technology, Ehitajate 5, 19086 Tallinn, Estonia

S. A. Kiselev and A. J. Sievers

Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14853-2501, USA

A theory is developed to describe the effect of an intrinsic localized mode (ILM) on small vibrations in a monatomic chain with hard quartic anharmonicity. One prediction is the appearance in the chain of linear local modes nearby the ILM. To check this result, molecular dynamics calculations of vibrations under strong local excitation are carried through with high precision. The results fully confirm the prediction.

I. INTRODUCTION

The realization[1–3] that there can exist stable strongly localized excitations in perfect classical anharmonic lattices has led to theoretical studies exploring a variety of possibilities.[4–12] The frequency of such a localized vibration depends on its amplitude and lies outside the phonon spectrum. Such excitations are called intrinsic localized modes (ILMs),[2] to emphasize their similarity in appearance to defect impurity modes, or called discrete breathers[7] or discrete solitons[13,14] to make a connection to solitons in continuous systems. Some experimental evidence for the existence of intrinsic localized modes in one-dimensional (1D) lattices in microscopic and macroscopic lattices has been demonstrated.[15–22]

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 backreaction on the phonon spectrum should have physical consequences since the ILM could induce local modes outside the plane wave spectrum. The presence of these additional resonances and their dependence on the amplitude of the ILM should add some complexity to the energy relaxation rate of the ILM to the phonon bath. In a different direction, since an ILM can move through the lattice it is expected that such trapped local modes would also move or, more likely, tend to inhibit the translational motion of such localized energy.

In this communication we examine analytically the small amplitude vibrations of a 1D nonlinear chain with intersite coupling in the presence of an ILM. The results show that an ILM with sufficient nonlinear amplitude stabilizes the appearance of linear local modes (LLMs) above the top of the plane wave spectrum. Next molecular dynamics (MD) simulations are used to verify these findings. First an ILM is formed and the power spectrum calculated, next the initial conditions are changed and very small amplitude shifts associated with a LLM are added to the neighboring atoms and the power spectrum calculated again. The cases when the new spectra contain one additional weak peak above the phonon spectrum are studied in detail. The frequencies of the additional peaks do not depend on their amplitudes but they

do depend on the ILM amplitude consistent with the linear requirement. Good agreement is obtained between the positions of the additional peaks and the theoretically calculated frequencies of the LLMs.

II. THEORY

Let us consider an anharmonic monatomic chain with interactions of nearest-neighboring atoms. Taking into account only quartic anharmonicity, the equation of motion of atoms in this model reads

$$\ddot{U}_n = \sum_{n'=n\pm1} [k_2(U_{n'} - U_n) + k_4(U_{n'} - U_n)^3], \quad (1)$$

where U_n are the reduced displacements of atoms located at the site n of the chain, the subscripts n indicate the number of the site, k_2 and k_4 are the parameters of harmonic and anharmonic springs, whereas $k_2 = \omega_m^2/4$ determines the top phonon frequency ω_m. If $k_4 > 0$ then ILMs may be excited in the chain with the frequencies above the allowed phonon spectrum.[2,4]

To describe small vibrations of the chain in presence of an ILM, we add to U_n an infinitesimal displacement q_n: $U'_n(t) = U_n(t) + q_n(t)$. For the displacements of the ILM we take $U_n(t) = A_n \cos \omega_L t$, where A_n is the amplitude parameter of the ILM which may be both, positive or negative (the small contribution of higher-order harmonics is omitted). The shift $U'_n(t)$ also satisfies Eq. (1). Subtracting from this equation the equation for $U_n(t)$ we get

$$\ddot{q}_n = \sum_{n'=n\pm1} [k_2 + 3k_4(A_n - A_{n'})^2 \cos^2 \omega_L t](q_{n'} - q_n). \quad (2)$$

One can divide q_n into two parts: (1) the shifts $q_{0,n}$ describing small variations of the ILM (they have been considered in Refs. 23 and 24) (2) all other shifts $q_{1,n}$. Here we are interested in the stable solutions of Eq. (2) which correspond to the latter modes. These modes are orthogonal to (i.e., independent from) the ILM. We can take the orthogonality condition into account if we add in Eq. (2) the term

$\lambda\Sigma_{n'}A_nA_{n'}q_{n'}$, where the Lagrange multiplier λ should be found from the orthogonality condition of the LLM under consideration. However, if one considers the linear modes of symmetry different from the ILM then there is no need to add the term $\propto\lambda$, since the orthogonality conditions are already fulfilled.

To identify LLMs, we first apply the rotating wave approximation which corresponds to the replacing in Eq. (2) the factor $\cos^2\omega_L t = (1+\cos 2\omega_L t)/2$ by $1/2$ [i.e., we neglect the oscillating in time term $(1/2)\cos 2\omega_L t$]. In this approximation the equation of motion of the linear modes takes the form

$$-\ddot{q}_n = \sum_{n'}(D_{nn'} + v_{nn'})q_{n'}, \qquad (3)$$

where $D_{nn'} = k_2(2\delta_{nn'} - \delta_{n\pm 1,n'})$ is the dynamical matrix of the perfect monatomic chain

$$v_{nn'} = \frac{3k_4}{2}\{\delta_{nn'}[(A_n - A_{n+1})^2 + (A_n - A_{n-1})^2] $$
$$- \delta_{n\pm 1,n'}(A_n - A_{n'})^2\} + \lambda A_n A_{n'} \qquad (4)$$

is the perturbation of the dynamical matrix.

The effect of the perturbation can be found by the Lifshitz method.[25] Thus a linear local mode l exists if the imaginary part of the Green's function of the perturbed lattice has a pole at ω_l outside the allowed phonon spectrum. The latter function can be found from the equation

$$G(\omega) = [I - G^{(0)}(\omega)v]^{-1}G^{(0)}(\omega), \qquad (5)$$

where G, v, and $G^{(0)}$ are matrices; v is given by Eq. (4),

$$G^{(0)}_{nn'}(\omega) = [-\rho(\omega)]^{|n-n'|}/\omega\sqrt{\omega^2 - 1} \qquad (6)$$

is the (n,n') component of the Green's function matrix of the perfect chain,[26] $\rho(\omega) = (\omega - \sqrt{\omega^2 - 1})^2 \leqslant 1$ (the units $\omega_m = 1$ are used). The amplitude parameters of a LLM are given by the relation

$$a_{n,l} = G_{nn_1}(\omega_l)/G_{n_1 n_1}(\omega_l). \qquad (7)$$

The time oscillatory terms in Eq. (2), neglected in the rotating wave approximation, can lead to effects not found in the vibrations of a lattice with a static defect. This difference follows from the Floquet theorem according to which in a periodically time-dependent system with the period T, besides the excitations with the frequency ω_l, there exist also excitations with the frequencies $|\pm\omega_l + 2\pi N/T|$, where N is an integer. In our case $T = \pi/\omega_L$; the repetitions of the frequency result from the time-dependent part of the anharmonic interaction of the LLMs with ILMs.

The time oscillatory terms in Eq. (2) can also cause a renormalization of the frequencies of linear modes. This follows from the fact that these terms oscillate in time with the frequencies $2\omega_L - \omega$ and $2\omega_L + \omega$ (here ω is the frequency of a small vibration), one of which, $2\omega_L - \omega_l$, may be comparable with the perturbation v. As a result, the frequencies of some linear modes may acquire complex values, which will cause these modes to become unstable.[27–30]

To find whether a LLM is stable or not we apply in Eq. (2) the expansion $q_n = \Sigma_j e_{nj}x_j$, where e_{nj} is the normalized contribution of the atom n to the normal coordinate x_j of the perturbed lattice. We get

$$-\ddot{x}_l = \omega_l^2 x_j + \cos 2\omega_L t \sum_{nn'}\sum_j e_{nl}w_{nn'}e_{n'j}x_j. \qquad (8)$$

The effect of the time oscillatory term in Eq. (8) is most important for the modes with frequencies close to ω_L. Supposing that only the LLM under consideration has such a frequency, we can neglect in the right-hand-side of this equation the terms $j \neq l$. In this approximation

$$-\ddot{x}_l = \omega_l^2(1 + h\cos 2\omega_L t)x_l, \qquad (9)$$

where $h = \omega_l^{-2}\Sigma_{nn'}e_{nl}w_{nn'}e_{n'l}$. [Here the inhomogeneous term $\propto x_L(t)\cos 2\omega_L t$ also has been neglected; this term describes the infinitesimal forced oscillations with the frequencies ω_L and $3\omega_L$ resulting in the infinitesimal shift of the ILM phase.] Equation (9) is the Mathieu equation describing the parametric resonance.[31] The term $\propto h$ may result in (a) renormalization of the frequency of the LLM, or (b) in the modes instability; in the latter case the renormalized frequency is complex. Below we consider the renormalization and the instability conditions numerically.

LLMs induced by even ILM

Both even and odd ILMs can exist in the nonlinear monatomic chain.[4] Here we consider an even ILM to be the effective defect in this lattice calculation, since this mode is stable with respect to small translational fluctuations.[28] This choice will permit calculations of ILMs+LLMs with high precision. The amplitude patterns for such an ILM for different values of the dimensionless nonlinear parameter, $k_4 A_0^2/k_2$, are given in rows 3 and 4 of Table I.

An even ILM can induce the appearance of both, odd and even LLMs. In the case of odd LLMs the λ multiplier in Eq. (4) equals zero. Then, if one considers an ILM localized of six neighboring central atoms, the perturbation matrix v [see Eq. (4)] is the six-range symmetric matrix with the following nonzero elements:

$$v_{00} = v_{11} = \gamma_0 + \gamma_1, \quad v_{01} = -\gamma_0, \quad v_{12} = -\gamma_1,$$

$$v_{22} = \gamma_1 + \gamma_2, \quad v_{23} = -\gamma_2, \quad v_{33} = \gamma_3, \qquad (10)$$

and with few matrix elements with negative index satisfying the relations $v_{-n-n'} = v_{n+1n'+1}$ $(n>0)$. Here $\gamma_n \approx 3k_4(A_n - A_{n+1})^2/2k_2$ are the changes of the springs (in dimensionless units). The effect of an even ILM upon even phonons is described by the perturbation matrix given by Eq. (4), which is equal to the previous perturbation matrix v plus the term $\lambda A_n A_{n'}$; the value of λ must be found self-consistently from the orthogonality condition $\Sigma_n e_{n,evn}A_n = 0$.

Inserting the matrix v into Eqs. (5) and (7) gives the frequencies and relative amplitudes of the LLMs. The parametric resonance parameter h, which determines the frequency correction equals

TABLE I. Analytical calculations of odd and even LLMs for a lattice containing an even ILM with a range of nonlinear parameter $k_4 A_0^2 / k_2$ values. Given are the relative ILM frequency ω_L / ω_m, the ILM amplitudes of the four next-to central particles A_n with $A_{-n} = -A_{n+1}$, the relative frequency of the odd and even LLM ω_{odd} / ω_m and ω_{evn} / ω_m, the amplitudes of these modes $a_{-n,odd} = a_{n+1,odd}$ and $a_{-n,evn} = -a_{n+1,evn}$ ($n \geqslant 0$); $A_1 - A_0 = 1$; $a_{1,odd} = a_{1,evn} = 1$. (The frequencies of the odd LLMs are corrected for the time oscillatory perturbation; h and h_{cr} are the corresponding parameters of this perturbation.)

$k_4 A_0^2 / k_2$	0.50	0.75	1.00	1.25	1.50	1.75	2.00
ω_L / ω_m	1.36	1.51	1.64	1.77	1.88	2.00	2.10
$-A_2$	0.199	0.170	0.153	0.141	0.133	0.127	0.121
A_3	0.042	0.027	0.019	0.015	0.012	0.010	0.008
ω_{odd} / ω_m	1.288	1.418	1.486	1.58	1.677	1.78	1.86
$a_{1,odd}$	0.729	0.784	0.819	0.844	0.862	0.876	0.887
$a_{3,odd}$	0.347	0.262	0.212	0.179	0.156	0.138	0.124
h_{odd}	0.456	0.560	0.629	0.679	0.711	0.746	0.770
$h_{odd,cr}$	0.52	0.63	0.77	0.84	0.89	0.94	0.98
ω_{evn} / ω_m	1.009	1.031	1.063	1.102	1.142	1.179	1.219
$a_{1,evn}$	0.383	0.308	0.273	0.252	0.238	0.228	0.220
$-a_{3,evn}$	1.143	0.822	0.639	0.522	0.442	0.384	0.340
$\tilde{\lambda} \cdot 10^2$	0.70	0.83	0.87	0.92	0.96	0.98	1.01

$$h = \frac{3 K_4 A_0^2}{K_2 \omega_l^2} \sum_{n \geqslant 1} (|e_n| + |e_{n+1}|)^2 (|A_n| + |A_{n+1}|)^2. \quad (11)$$

In Table I the calculated values for odd and even LLMs are presented for different values of the nonlinear parameter of the ILM, $k_4 A_0^2 / k_2$. Comparing the LLM amplitude patterns with the amplitudes of the ILM shows that they are mutually orthogonal and the ILM and the LLM belong to the different degrees of freedom. Note that the frequency of the odd LLM is rather close to ω_L. For this mode consideration of the time oscillatory term in Eq. (9) is important. The numerical calculations of the Mathieu equation (9) shows that in all cases the parametric resonance parameter h is less than its critical value h_{cr} for the instability. The frequency of the even LLM is significantly different from ω_L so that the correction caused by the time oscillatory term is now small.

III. MD SIMULATIONS

To perform MD simulations of ILMs and LLMs, we integrate numerically the equation of motion given by Eq. (1). We calculate the vibrational amplitudes and velocities of atoms of the finite chain with 10^4 atoms in 10^5 time points. The time interval includes $\sim 10^3$ periods of the ILMs. We also calculate the vibrational power spectrum. To decrease the background in the spectrum, Fourier transformations are carried out between the time points corresponding to the same phase of vibrations. For the initial condition, we used the ILM displacements of the six central atoms from their equilibrium positions. The amplitude difference of the two central atoms is fixed at unity with $k_2 = 100$ ($\omega_m = 20$). In this case the actual values of k_4 for ILMs with frequency $\leqslant 2\omega_m$ are of the order of k_2. The initial shifts of other four central atoms are selected so that only the ILM is excited. The re-

sulting amplitude parameters A_n of six central atoms are those shown in Table I.

Examples of the calculated power spectrum of an even ILM are given in the bottom panels of Fig. 1. Two different values of the nonlinear parameter are shown. Each spectrum
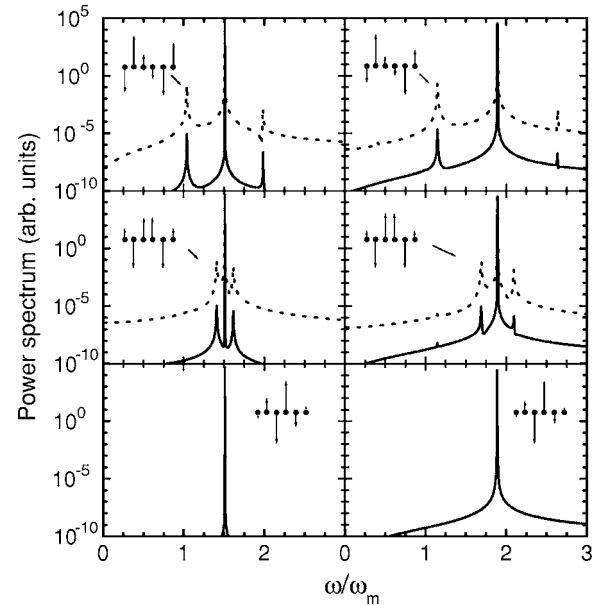

FIG. 1. Power spectra of ILMs for two different nonlinear parameter values with and without LLMs. Left column of figures: the main spectral feature is an even ILM with $k_4 A_0^2 / k_2 = 0.75$, right column of figures: same even ILM but with $k_4 A_0^2 / k_2 = 1.5$. Bottom panels: the unperturbed even ILM; middle panels: an ILM perturbed by an odd LLM; top panels: an ILM perturbed by an even LLM. Although the dotted and solid lines differ by 10^2 in the LLM amplitude its frequency remains unchanged. The amplitude patterns of the ILMs and LLMs are shown by arrows.

has a single peak with a frequency $\omega_L > \omega_m$. Not shown are the much weaker peaks associated with the higher odd harmonics at $3\omega_L$, $5\omega_L$, etc. Hence, the bottom panels demonstrate that our initial conditions correspond solely to the excitation of an ILM (at least with the accuracy 10^{-14}).

When small additional amplitude shifts appropriate to the odd or even symmetry LLM presented in Table I are added to the ILM amplitude pattern, then additional small peaks appear in the power spectrum, one at $\omega < \omega_L$, and an even smaller symmetrically situated peak at $\omega > \omega_L$. The middle panels in Fig. 1 show the case for an odd LLM, with its amplitude pattern, and the top panel for an even LLM, again with its amplitude pattern. One finds analogous sideband spectra for other $k_4 A_0^2/k_2$ values. These data show that the frequencies of the spectral peaks depend on the amplitude and, hence, the frequency of the ILM. At the same time the solid and dotted traces presented in Fig. 1, where the LLM amplitude is varied by 10^2, demonstrate that these LLM frequencies do not depend on their own amplitudes as expected for a linear response (the amplitudes of the LLMs are $\sim 10^5$ and $\sim 10^3$ times less than the amplitude of the ILM). The $\tilde{\omega}_l = 2\Omega_L - \omega_l$ of the symmetrically situated weak peak indicates that this spectral feature is the four wave mixing response of the LLM with the ILM.

IV. DISCUSSION AND CONCLUSIONS

There is value in a quantitative comparison of the analytical results and MD simulations for the LLMs. Figure 2 presents the frequencies of the LLMs obtained by both these methods.

Inspection of these results shows that there is excellent agreement between the two methods. Note that the time oscillatory terms provide a significant correction for odd LLM case but are insignificant for the even LLM. The reason is in the relatively small difference of $\omega_L - \omega_{odd}$ as compared to $\omega_L - \omega_{evn}$.

We have presented a theory which allows one to describe the effect of an ILM on phonons in a nonlinear monatomic chain. The prediction of linear localized modes above the top of the plane wave spectrum is in good agreement with MD simulations. Basically the appearance of the ILM changes the nearby nonlinear spring constants sufficiently so that linear localized modes can also appear. The resulting lattice perturbation produced by the ILM is analogous to that associated with a force constant defect in a linear lattice in that a
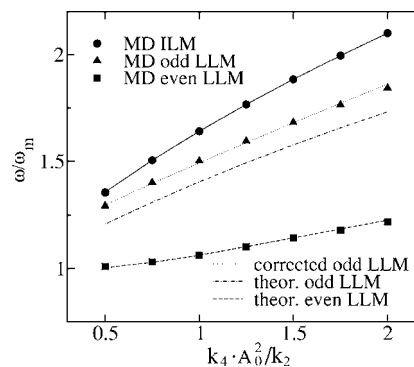

FIG. 2. The dependence of the frequencies of the odd and even LLMs on the dimensionless nonlinear parameter $k_4 A_0^2/k_2$ of the ILM. The frequencies of the odd LLM are given with (dotted line) and without (dash-dotted line) taking into account the time-oscillatory term $h \cos 2\omega_l t$ in Eq. (9). The ILM frequency dependence is also shown.

small number of degrees of freedom can be strongly perturbed but all phonons are perturbed to some extent. Looking ahead to more realistic diatomic lattices involving two body potentials,[9] we expect that a variety of LLM possibilities may appear. These could include modes of the local, gap, and resonant types as well as tunneling states, all made possible by the fundamental combination of nonlinearity and lattice discreteness. One may anticipate that, in analogy with the previously studied force constant defect cases, ILM-induced IR and Raman activity may occur throughout the entire phonon spectrum.[32,33]

There are also some very interesting differences between the properties of extrinsic and intrinsic localized excitations evident from our study of this simple model system. One is that an ILM introduces a periodically time-dependent perturbation that supports nonlinear mixing processes between the ILM and the LLMs and a second is that for a moving ILM the "effective defect space" with its associated LLMs will travel together with the nonlinear excitation.

ACKNOWLEDGMENTS

*Email address: shell@fi.tartu.ee

[1] A. S. Dolgov, Fiz. Tverd. Tela (Leningrad) **28**, 1641 (1986) [Sov. Phys. Solid State **28**, 907 (1986)].

[2] A. J. Sievers and S. Takeno, Phys. Rev. Lett. **61**, 970 (1988).

[3] S. Takeno, K. Kisoda, and A. J. Sievers, Prog. Theor. Phys. Suppl. **94**, 242 (1988).

[4] J. B. Page, Phys. Rev. B **41**, 7835 (1990).

[5] S. A. Kiselev, S. R. Bickham, and A. J. Sievers, Comments Con-

dens. Matter Phys. **17**, 135 (1995).

[6] A. J. Sievers and J. B. Page, in *Dynamical Properties of Solids: Phonon Physics The Cutting Edge*, edited by G. K. Norton and A. A. Maradudin (North Holland, Amsterdam, 1995), Vol. VII, p. 137.

[7] S. Flach and C. R. Willis, Phys. Rep. **295**, 181 (1998).

[8] R. Lai and A. J. Sievers, Phys. Rep. **314**, 147 (1999).

[9] S. A. Kiselev, S. R. Bickham, and A. J. Sievers, Phys. Rev. B **48**,

13508 (1993).

[10] U. Schroder, D. Bonart, and A. P. Mayer, Physica B **220**, 390 (1996).

[11] S. A. Kiselev and A. J. Sievers, Phys. Rev. B **55**, 5755 (1997).

[12] H. Fehske, M. Kinateder, G. Wellein, and A. R. Bishop, Phys. Rev. B **63**, 245121 (2001).

[13] M. Toda, *Theory of Nonlinear Lattices*, Springer Series in Solid State Sciences, 2nd ed. (Springer-Verlag, Berlin, 1989), p. 23.

[14] D. N. Christodoulides and R. I. Joseph, Opt. Lett. **13**, 794 (1988).

[15] B. I. Swanson, J. A. Brozik, S. P. Love, G. F. Strouse, A. P. Shreve, A. R. Bishop, W. Z. Wang, and M. I. Salkola, Phys. Rev. Lett. **82**, 3288 (1999).

[16] U. T. Schwarz, L. Q. English, and A. J. Sievers, Phys. Rev. Lett. **83**, 223 (1999).

[17] E. Trias, J. J. Mazo, and T. P. Orlando, Phys. Rev. Lett. **84**, 741 (2000).

[18] P. Binder, D. Abraimov, A. V. Ustinov, S. Flach, and Y. Zolota-ryuk, Phys. Rev. Lett. **84**, 745 (2000).

[19] M. Sato, B. E. Hubbard, A. J. Sievers, B. Ilic, D. A. Czaplewski, and H. G. Craighead, Phys. Rev. Lett. **90**, 044102 (2003).

[20] J. W. Fleischer, M. Segev, N. K. Efremidis, and D. N. Christodoulides, Nature (London) **422**, 147 (2003).

[21] M. Sato and A. J. Sievers, Nature (London) **342**, 486 (2004); Phys. Rev. B **71**, 214306 (2005).

[22] J. P. Wrubel, M. Sato, and A. J. Sievers, Phys. Rev. Lett. **95**, 264101 (2005).

[23] D. Nevedrov, V. Hizhnyakov, and A. J. Sievers, NATO Sci. Ser.: II: Math. Phys. Chem. **39**, 343 (2001); V. Hizhnyakov, D. Nevedrov, and A. J. Sievers, Physica B **316-317**, 132 (2002).

[24] V. Hizhnyakov, A. Shelkan, and M. Klopov (unpublished).

[25] A. A. Maradudin, E. W. Montroll, G. S. Weiss, and I. P. Ipatova, in *Theory of Lattice Dynamics in the Harmonic Approximation*, Solid State Physics Supp. 3, 2nd ed., edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic Press, New York, 1971); A. A. Maradudin, in *Theoretical and Experimental Aspects of the Effects of Point Defects and Disorder of the Vibrations of Crystals*, Solid State Physics, edited by F. Seitz and D. Turnbull (Academic Press, New York, 1966), Vols. 18 and 19.

[26] E. N. Economou, *Green's Functions in Quantum Physics* (Springer-Verlag, Berlin, 1983).

[27] Y. S. Kivshar and M. Peyrard, Phys. Rev. A **46**, 3198 (1992).

[28] K. W. Sandusky, J. B. Page, and K. E. Schmidt, Phys. Rev. B **46**, 6161 (1992).

[29] I. Daumont, T. Dauxois, and M. Peyrard, Nonlinearity **46**, 617 (1997).

[30] R. Lai and A. J. Sievers, Phys. Rev. B **57**, 3433 (1998).

[31] L. D. Landau and E. M. Lifshiz, *Mechanics* (GIFML, Moscow, 1958) (see paragraph "Parametric resonance").

[32] A. S. Barker and A. J. Sievers, Rev. Mod. Phys. **47**, S1 (1975).

[33] M. Cardona and M. L. W. Thewalt, Rev. Mod. Phys. **77**, 1173 (2005).