

Second-harmonic generation in diatomic lattices

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It is shown that in a diatomic lattice with cubic nonlinearity resonant second-harmonic generation can take place. Conditions for the phenomenon to be observed are established. Systems of equations governing three- and two-wave interactions in a lattice with a complex cell are deduced. [S1063-651X(96)05509-2]

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I. INTRODUCTION

Propagation of a small-amplitude envelope soliton in any lattice with cubic nonlinearity is necessarily accompanied by a harmonic having a frequency two times greater than the frequency of the discrete carrier wave (cw) [1,2]. In a one-dimensional monatomic lattice this process has nonresonant character. This means that the double frequency excitations are localized about the soliton and have amplitudes much smaller than the soliton one (such terms will be referred to as companion terms, while the leading order solution will be called a principal term). Suppose now that a one-dimensional multiautomic lattice is under consideration and that the cw belongs to the acoustic branch. Under definite conditions it may occur that the duplicated cw frequency lies on one of the optical branches. Then propagation of linear waves with such a frequency is allowed and the process of energy transformation from the principal term to the companion one becomes resonant. Using the terminology accepted in nonlinear optics [3] one can say that this situation corresponds to the second-harmonic generation.

The process of the second-harmonic generation in optical media is well studied [3,4]. In particular, it is known that there are two important conditions for the phenomenon to be observed. The first one is the presence of nonlinearity which, as has been mentioned above, results in doubling the cw frequency. The other important factor is the dispersion which prevents appearing higher harmonics. Both these factors are intrinsic properties of a lattice with cubic nonlinearity. Meanwhile, there are some specific features of the second-harmonic generation in lattices. The most important one is that the discreteness introduces effective periodicity and that is why the so-called synchronism condition takes a special form. The second feature is that not any lattice with a complex cell allows the second-harmonic generation and some requirements on the lattice parameters must be imposed. In particular, the phenomenon is observed only for a definite cw wavelength and the number of modes originating the second-harmonic generation is related to the number of atoms in the cell. At last, the process is accompanied by a nonresonant excitation of companion terms.

As is known from the soliton theory [4,5], the second-harmonic generation is described by a system of equations

which can be treated as a degenerate case of a system governing resonant three-wave interactions. That is why in the present paper we first derive equations for three-wave interactions in a lattice with a complex cell, and then analyze them for the case of a diatomic lattice when two modes coincide.

The paper is organized as follows. In Sec. II using the technique developed in [1] the systems governing the three-wave interaction and the second-harmonic generation are derived. Examples of the second-harmonic generation in diatomic lattices are considered in Sec. III. The results are summarized in the Conclusion.

II. EVOLUTION EQUATIONS

To be more specific we consider a lattice described by the Hamiltonian of a rather general form

$$\begin{aligned}
 H = & \frac{1}{2} \sum_{n,\alpha} M_\alpha [\dot{u}_\alpha(n)]^2 \\
 & + \frac{1}{2} \sum_{n_1,\alpha_1} \sum_{n_2,\alpha_2} K_2(n_1,\alpha_1; n_2,\alpha_2) u_{\alpha_1}(n_1) u_{\alpha_2}(n_2) \\
 & + \frac{1}{3} \sum_{n_1,\alpha_1} \dots \sum_{n_3,\alpha_3} K_3(n_1,\alpha_1; \dots; n_3,\alpha_3) \\
 & \times u_{\alpha_1}(n_1) \dots u_{\alpha_3}(n_3). \tag{1}
 \end{aligned}$$

Here $u_\alpha(n)$ is a small displacement from the equilibrium position of the α th atom in the n th cell, M_α is a mass of the α th atom, $K_p(n_1,\alpha_1; \dots; n_p,\alpha_p)$ are linear ($p=2$) and nonlinear ($p=3$) force constants, and the overdot stands for the derivative with respect to time.

For the mathematical treatment of the phenomenon it is convenient to introduce the terminology as follows [6]. The pair (n,α) will be designated as x : $x=(n,\alpha)$, the displacement of an atom will be described by a normalized function $v(x)$: $v(x) = \sqrt{M_\alpha} u_\alpha(n)$, and interactions among the atoms will be characterized by

$$J_p(x_1; \dots; x_p) = \frac{K_p(n_1,\alpha_1; \dots; n_p,\alpha_p)}{\sqrt{M_{\alpha_1} \dots M_{\alpha_p}}}.$$

The coefficients $J_p(x_1; \dots; x_p)$ are invariant with respect to permutations of the arguments. Then the dynamical equation describing the lattice reads

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$$\ddot{v}(x) = - \sum_{x_1} J_2(x; x_1) v(x_1) - \sum_{x_1, x_2} J_3(x; x_1; x_2) v(x_1) v(x_2). \quad (2)$$

In what follows the consideration will be restricted to the case of a homogeneous lattice when

$$J_p(x_1; \dots; x_p) = J_{\alpha_1, \dots, \alpha_p}(n_1 - n_2; \dots; n_{p-1} - n_p). \quad (3)$$

We are concerned with solutions of Eq. (2) characterized by the amplitude of oscillations A being much less than the relation $|K_2/K_3|$. This determines the effective small parameter of the problem which can be introduced as $\mu = A \max |K_3/K_2| \ll 1$, and allows one to employ the method of multiscale expansion [7] against the discrete cw background (see, e.g., [8,2,1]). This means that the solution of Eq. (2) is represented in a form of a series $v(x) = \sum_{\nu=1}^{\infty} \mu^{\nu} v_{\nu}(x)$. The small parameter μ defines also a set of ‘‘times’’ $t_{\nu} = \mu^{\nu} t$, which are regarded as independent variables, and therefore $d/dt = \sum_{\nu=0}^{\infty} \mu^{\nu} (\partial/\partial t_{\nu})$, and a set of spatial coordinates $n_{\nu} = \mu^{\nu} n$. Subject to rather general conditions, established in [1], all the spatial variables with $\nu \geq 1$ can be treated as continuum ones. More precisely, we introduce slow coordinates $X_{\nu} = a n_{\nu}$ ($\nu \geq 1$), where a is a lattice constant, and hold the notation X_0 for the pair of discrete variables (n_0, α) .

Substitution of the expansion for $v(x)$ into the dynamical equation (2) in the leading order results in the equation

$$\frac{\partial^2 v_1(X_0, t_0)}{\partial t_0^2} + \sum_{X'_0} J_2(X_0; X'_0) v_1(X'_0, t_0) = 0. \quad (4)$$

Hereafter only the most ‘‘rapid’’ arguments of the dependent variables are indicated [so, for example, $v_1(X_0, t_0)$ depends on all X_{ν} and t_{ν}]. Equation (4) is associated with the eigenvalue problem

$$\omega_{\gamma, q}^2 \phi_{\gamma, q}(x) = \sum_{x_1} J_2(x; x_1) \phi_{\gamma, q}(x_1). \quad (5)$$

Here $\omega_{\gamma, q}$ can be interpreted as a frequency of a mode characterized by a pair of quantum numbers (γ, q) , which label a branch of the spectrum (γ) and a wave number (q) inside the first Brillouin zone (BZ). For the sake of definiteness we use $\gamma = 1$ for the acoustic branch and $\gamma \geq 2$ for optical branches. It is assumed that the eigenvalue problem (5) is subject to the cyclic boundary conditions, i.e., that the lattice is finite and has \mathcal{N} cells. After all we are interested in the limit $\mathcal{N} \rightarrow \infty$, which allows us to calculate all quantities with the accuracy $O(\mathcal{N}^{-1})$. The eigenfunctions $\phi_{\gamma, q}(x)$ are chosen to make up a complete orthonormal set:

$$\begin{aligned} \sum_x \bar{\phi}_{\gamma_1, q_1}(x) \phi_{\gamma_2, q_2}(x) &= \delta_{\gamma_1 \gamma_2} \delta_{q_1 q_2}, \\ \sum_{\gamma, q} \bar{\phi}_{\gamma, q}(x) \phi_{\gamma, q}(x_1) &= \delta_{x x_1} \end{aligned} \quad (6)$$

(δ being the Kronecker delta and the bar standing for the complex conjugation).

The last property allows one to represent a general three-mode solution in the form

$$v_1(X_0, t_0) = \sum_{j=1}^3 A_j(X_1, t_1) e^{-i\omega_j t_0} \phi_{\gamma_j, q_j}(X_0) + \text{c.c.}, \quad (7)$$

where $A_j(X_1, t_1)$ are slowly varying amplitudes and ω_j stands for ω_{γ_j, q_j} .

In the second order of μ one obtains

$$\begin{aligned} \frac{\partial^2 v_2(X_0, t_0)}{\partial t_0^2} + \sum_{X'_0} J_2(X_0; X'_0) v_2(X'_0, t_0) \\ = -2 \frac{\partial^2}{\partial t_0 \partial t_1} v_1(X_0, t_0) \\ - \sum_{X'_0} a(n'_0 - n_0) J_2(X_0; X'_0) \frac{\partial}{\partial X_1} v_1(X'_0, t_0) \\ - \sum_{X'_0, X''_0} J_3(X_0; X'_0; X''_0) v_1(X'_0, t_0) v_1(X''_0, t_0). \end{aligned} \quad (8)$$

Taking into account the explicit form (7) one concludes that the companion term may contain only the frequencies ω_j and $\omega_j \pm \omega_k$ with $j, k = 1, 2, 3$. The modes with ω_j come from the first two terms on the right-hand side of Eq. (8) and constitute a peculiarity of a multiatomic lattice since they are excited due to the existence of different branches of the spectrum and disappear in the case of a monatomic lattice [1].

By analogy with the leading order, $v_2(X_0, t_0)$ can be represented as an expansion over the eigenmodes $\phi_{\gamma, q}(X_0)$ which take into account all composite frequencies mentioned above. The coefficients of such an expansion are obtained by applying $\sum_{X_0} \bar{\phi}_{\gamma, q}(X_0)$ to both sides of Eq. (8). In this way one finds that the effect of the nonlinearity is described by

$$\begin{aligned} \mathcal{J}_{\gamma_k, \gamma_l, \gamma_m}(q_k, q_l, q_m) &= \sum_{x_1, x_2, x_3} J_3(x_1; x_2; x_3) \bar{\phi}_{\gamma_k, q_k}(x_1) \\ &\times \phi_{\gamma_l, q_l}(x_2) \bar{\phi}_{\gamma_m, q_m}(x_3). \end{aligned} \quad (9)$$

This sum can be transformed taking into account property (3) and the general form of the eigenmodes

$$\phi_{\gamma, q}(x) = \frac{1}{\sqrt{\mathcal{N}}} a_{\gamma, q}(\alpha) e^{iqan}, \quad (10)$$

where $a_{\gamma, q}(\alpha)$ is a normalized complex amplitude of oscillations of an atom α . The result reads

$$\begin{aligned} \mathcal{J}_{\gamma_k, \gamma_l, \gamma_m}(q_k, q_l, q_m) \\ = \frac{1}{\sqrt{\mathcal{N}}} \delta_{q_l - q_m - q_k, Q} \\ \times \sum_{\alpha_1, \alpha_2, \alpha_3} \bar{a}_{\gamma_k, q_k}(\alpha_1) a_{\gamma_l, q_l}(\alpha_2) \bar{a}_{\gamma_m, q_m}(\alpha_3) \\ \times \sum_{p, n} e^{i(pq_m - nq_k)} J_{\alpha_1, \alpha_2, \alpha_3}(n, p). \end{aligned} \quad (11)$$

known as the normal (N) and umklapp (U) process. The difference between these situations becomes important when one considers kinetic properties related to phonon-phonon interaction [9]. In the case at hand we also find qualitative distinction between N and U processes. Indeed, $v_1 v_2 < 0$ if $\tilde{q} < q_1 < \pi/(2a)$, where $\tilde{q} \approx 1.4989a^{-1}$ is the smallest possible value of q_1 which is reached at $M_1 \rightarrow M_2$, $\mathcal{K}_1 \rightarrow \mathcal{K}_2$, and $Q = 0$. This means that in the mentioned region of the wave numbers *the second-harmonic generation is accompanied by a change of the direction of the energy flux*. Otherwise, when $\pi/(2a) < q_1 < \pi/a$, the synchronism conditions are satisfied at nonzero Q and $v_1 v_2 > 0$. Thus the peculiarity of our case is that weak nonlinearity results in a change of the group velocity to opposite in an N process while in a U process the sign of the velocity is preserved.

Resonant two-wave interactions are sensitive to the type of nonlinearity. So, for example, in the case of a cubic on-site potential $K_3(x; x_1; x_2) = W_\alpha \delta_{xx_1} \delta_{xx_2}$ (W_α characterizes the intensity of the nonlinear force) the lattice is governed by the Hamiltonian

$$H = \sum_n \left\{ \frac{1}{2} M_1 [\dot{u}_1(n)]^2 + \frac{1}{2} M_2 [\dot{u}_2(n)]^2 + \frac{1}{2} \mathcal{K}_1 [u_1(n) - u_2(n-1)]^2 + \frac{1}{2} \mathcal{K}_2 [u_2(n) - u_1(n)]^2 + \frac{1}{3} W_1 [u_1(n)]^3 + \frac{1}{3} W_2 [u_2(n)]^3 \right\}. \quad (19)$$

From Eq. (11) one calculates

$$\mathcal{J}_{1,2,1}(q_1, q_2, q_1) = \frac{1}{\sqrt{\mathcal{N}}} \sum_{\alpha=1,2} \frac{W_\alpha}{M_\alpha^{3/2}} \bar{a}_{1,q_1}^2(\alpha) a_{2,q_2}(\alpha). \quad (20)$$

Thus, the effective nonlinearity depends on a wave number only through the amplitudes of oscillations, $a_{\gamma,q}(\alpha)$, of each atom. In the case of nonlinear intersite interactions the coefficient $\mathcal{J}_{1,2,1}(q_1, q_2, q_1)$ becomes explicitly dependent on the wave numbers. A physical reason for this is quite transparent: in the last case the effective nonlinear interaction depends on the phase mismatch between neighbor particles.

It is to be emphasized that the requirement (18) is satisfied in the limit $\mathcal{K}_2 \rightarrow \mathcal{K}_1$, $M_1 \rightarrow M_2$. This, however, does not mean ‘‘analytical’’ transition to a monatomic lattice. The existence of *two* branches of the spectrum is crucial for the possibility of the second-harmonic generation. In order to illustrate this we note that in the above limit $q_1 \rightarrow \tilde{q} < \pi/2a$, while the frequency of the second harmonic tends to $\tilde{\omega} \approx 1.035\omega(\pi/a)$.

In the opposite limit when

$$\sqrt{\mathcal{K}_1 \mathcal{K}_2 M_1 M_2} = \frac{\sqrt{3}}{8} (\mathcal{K}_1 + \mathcal{K}_2) (M_1 + M_2),$$

$$q_l = \frac{\pi}{a}, \quad q_m = 0, \quad Q = -\frac{2\pi}{a}, \quad (21)$$

the gap has a maximal width and the energy transforms from the standing acoustic wave at the BZ edge to the optical mode with the top frequency $\omega_2 = \omega_{2,0} = \sqrt{(\mathcal{K}_1 + \mathcal{K}_2)(M_1 + M_2)/(M_1 M_2)}$. In this case $v_1 = v_2 = 0$ and the solution of Eqs. (16a) and (16b) describing the process in which the energy initially is concentrated in the acoustic mode reads (see, e.g., [3])

$$A_1 = \mathcal{A} \operatorname{sech}\left(\frac{t_1}{\tau}\right), \quad A_2 = \frac{1}{2i} \mathcal{A} \tanh\left(\frac{t_1}{\tau}\right). \quad (22)$$

Here \mathcal{A} is the initial amplitude of the acoustic wave and the absolute value of τ , where

$$\frac{1}{\tau} = \frac{\mathcal{A}}{2C\sqrt{\mathcal{K}_1 + \mathcal{K}_2}(M_1 + M_2)} \left\{ W_1 \frac{M_2}{M_1} \left[\frac{M_2 - M_1}{M_2 + M_1} - C \right] - W_2 \frac{M_1}{M_2} \left[\frac{M_2 - M_1}{M_2 + M_1} + C \right] \right\} \quad (23)$$

($C = \sqrt{1 - \sqrt{3}/2}$) plays the part of the effective time of the energy transfer from the acoustic mode to the optical one.

Notice that $\tau \rightarrow \infty$ as $W_1 \rightarrow W_2$ and $M_2 \rightarrow M_1$ (i.e., when the optical branch of the spectrum appears only due to different linear forces $\mathcal{K}_{1,2}$ among the atoms). This means that the second-harmonic generation does not occur. The physical origin of the phenomenon lies in the fact that the effective energy of nonlinear interaction between two modes belonging to different branches goes to zero in the above limit [what can be easily verified by direct calculation of the last two terms in Eq. (19)].

A common property of the phenomena of two- and three-wave interactions in a multiatomic lattice is excitation of a companion term. Taking into account the diversity of frequencies generated due to the nonlinearity, the companion term has a rather cumbersome form to be represented here for the generic case. That is why $v_2(X_0, t_0)$ is given below only for the case of the second-harmonic generation in a diatomic lattice (17). In this situation one calculates from Eq. (8)

$$v_2(X_0, t_0) = -\frac{2}{\omega_{2,0}} |A_2|^2 \mathcal{J}_{2,2,2}(0, q_2, q_2) \phi_{2,0}(X_0) + \frac{\Gamma_{2,1}(q_1, q_1)}{\omega_1^2 - \omega_{2,q_1}^2} \frac{\partial A_1}{\partial X_1} e^{-i\omega_1 t_0} \phi_{2,q_1}(X_0) + \frac{\Gamma_{2,1}(q_2, q_2)}{\omega_2^2 - \omega_{2,q_2}^2} \frac{\partial A_2}{\partial X_1} e^{-i\omega_2 t_0} \phi_{1,q_2}(X_0) + \sum_{\gamma=1,2} \left\{ 2A_1 A_2 \frac{\mathcal{J}_{1,\gamma,2}(q_1, 3q_1, q_2)}{\omega_{\gamma,3q_1}^2 - [3\omega_1]^2} e^{-3i\omega_1 t_0} \phi_{\gamma,3q_1}(X_0) + A_2^2 \frac{\mathcal{J}_{2,\gamma,2}(q_2, 2q_2, q_2)}{\omega_{\gamma,4q_1}^2 - [4\omega_1]^2} e^{-4i\omega_1 t_0} \phi_{\gamma,4q_1}(X_0) \right\} + \text{c.c.} \quad (24)$$

Here all wave numbers $p q_{1,2}$ (p being an integer) must be understood in the generalized sense $p q_{1,2} + Q$ with a vector of the reciprocal lattice Q chosen in such a manner that the resultant belongs to the first BZ.

The last formula has a number of important consequences. First, it follows from Eq. (24) that in a generic case the cubic potential results in a constant displacement (it is described by the first term in the right-hand side). This *displacement is determined by the optical branch* of the spectrum. The next two terms in the right-hand side of Eq. (24) mean that the *second harmonic is excited also in a nonresonant way*. They correspond to eigenmodes having the same wave numbers as the principal modes do but belonging to different branches of the spectrum as shown in Fig. 1 (this is a general peculiarity of multiautomic lattices [1]). The amplitudes of these companion modes are determined by spatial variations of the envelopes of the principal term and disappear when energy transforms between two nonmodulated waves. At last, Eq. (24) displays nonresonant excitation of the third and quartic harmonics (the last two terms in the right-hand side).

IV. CONCLUSION

To conclude, it has been shown that in multiautomic lattices with cubic nonlinearity resonant interactions among harmonics may take place. The systems of equations governing the three-wave process and the second-harmonic generation were derived. Though a second harmonic is generated in any diatomic lattice with cubic nonlinearity *the phenomenon has resonant character only if the gap of the spectrum is not more than $\omega_{2,0}(\sqrt{1+C}-\sqrt{1-C})/\sqrt{2}$* . The second-harmonic generation occurs at definite frequencies of the cw and is accompanied by modes excited in a nonresonant way. Meanwhile, the particular solution (22) represented here does not exhaust the diversity of effects described by the systems (16a) and (16b). So, for instance, using the analogy with nonlinear optics one can predict amplification of an acoustic mode by an optical one, periodic energy exchange between the branches, etc.

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