Nonlinear phonon generation via localized modes

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A theoretical study of parametric excitation of phonons involving impurities in cubic crystals with NaCl structures is presented. The proposed excitation mechanism which takes place via nonlinear processes involving localized modes is analogous to the first-order Suhl instability in ferromagnetic insulators. Sufficient excitation of impurity lattice vibration by infrared absorption can result in a sudden avalanche of optical and acoustic phonons due to anharmonic interaction. It is shown that the critical power of the laser source necessary to attain the instability threshold is experimentally feasible.

I. INTRODUCTION

Orbach[1] was the first one to draw attention to the fact that the decay of a long-wave optical phonon into two short-wave acoustic phonons is a parametric process, and therefore the generation of optical phonons using light might produce an instability in the system of acoustic phonons. The instability threshold should be especially low for the decay into transverse acoustic phonons whose lifetime is anomalously long.[2]

With the development of highly intense radiation sources in the microwave, infrared, and optical regions, the study of the nonlinear interaction of electromagnetic radiation with semiconductors[3] and plasmas[4] has stimulated considerable theoretical and experimental interest. One important area is the amplification of optical phonons[5] and ultrasound in semiconductors under intense laser radiation. This amplification can be attributed to the absorption of the laser radiation by conduction electrons, which is accompanied by either emission or absorption of phonons. The effect of a strong magnetic field has been also considered in these parametric processes.[7,8]

It is well known that when a substitutional impurity atom is introduced in a crystal, it gives rise to certain changes in the vibrational spectrum of the system.[9] In particular, if the impurity is lighter and/or the force constant is greater than that of the host crystal atoms, then a so-called localized mode appears above the quasicontinuum of phonons. The characteristics of this localized mode are that when it is excited, only the atoms near the impurity participate in the motion (the attenuation length decreases as the frequency increases); and that its frequency is higher than the maximum phonon frequency. The latter may be looked upon as the reason why the mode is localized; its frequency is too high to propagate in the lattice. The localized mode is a single mode of the system with a sharp frequency.

It is the purpose of this paper to present a simple theoretical study of nonlinear phonon generation involving localized modes associated with impurities in cubic crystals with NaCl structures. Here we are concerned with first-order nonlinear processes due to anharmonic interaction, i.e., three interacting phonon modes such that one is a localized mode. Physically, the proposed excitation mechanism is the following: The anharmonic interaction in cubic crystals containing impurities can induce transitions between localized modes and the host phonon wave modes. So, in analogy to the well-known Suhl instability which occurs at high power levels in ferromagnetic resonances, if a localized mode is pumped by a strong electromagnetic field, one finds that beyond a critical value of the field, the phonon wave modes, directly coupled to the pumped localized mode, grow parametrically, causing instabilities. When this condition is satisfied, the growth rate from pumping exceeds the decay rate from the various relaxation processes.

II. THEORETICAL APPROACH

In this section we shall derive an asymptotic expression for the phonon wave amplitudes associated with the *s*-like localized modes which will be used to treat the anharmonic effects.

We first consider a simple NaCl-type cubic lattice composed of N ions in which each ion in the crystal interacts only with its nearest neighbors. This nearest-neighbor interaction model is greatly oversimplified because it has the physically unrealistic feature that the components of the displacement vector in the direction of the three principal components are not coupled. However, this model has the advantage that it is simple enough for many of its properties to be studied analytically rather than numerically.[10] Let us suppose that a point imperfection is located at the origin (0,0,0), and for the other sites, host atoms are arranged such that $M_m = M_1$ for m even and $M_m = M_2$ for m odd where $m = m_1 + m_2 + m_3$ and (m_1, m_2, m_3) is a set of three integers denoting the position of the ions. Within this approach Takeno[11] has found that there exist three types of vibrational modes associated with the motion of the impurity. Among these modes, we are particularly interested in the solutions of the following equation:

$$D(\omega_s)=1+\xi-(\xi+\epsilon)M_1\epsilon_s^2\mu_s G(\omega_s^2;0,0,0)$$
$$+\xi(1-\epsilon)M_1\epsilon_s^2 G(\omega_s^2;1,0,0)=0\ , \tag{2.1}$$

which are directly associated with the motion of the impurity atom characterized by a large amplitude (localized s mode) predominantly located at the impurity and thus excitable by an external radiation. The remaining, the p-like (threefold degenerate) and the d-like (twofold degenerate) modes have a node at the site of the impurity, and therefore they are infrared inactive modes. In Eq. (2.1) the impurity parameters are defined as $\xi=(\phi'/\phi)-1$ and $\epsilon=1-(M_1'/M_1)$, where ϕ and M_1 are the force constant and the ion mass, respectively, of the pure crystal and M_1' is the mass of the substitutional impurity atom which interacts with its nearest-neighbors with the force constant ϕ'; and $G(\omega_s^2;m_1,m_2,m_3)$ is the Green function which is given by

$$G(\omega_s^2;m_1m_2m_3)=\frac{1}{N}\sum_{\mathbf{q}}\frac{\exp[i(q_1m_1+q_2m_2+q_3m_3)]}{M^*\omega_s^2-M^*\omega_q^2}\ , \tag{2.2}$$

where $M^*\omega_q^2$ determines the eigenfrequencies of the unperturbed crystal and $M^*\omega_s^2$ is defined by

$$M^*\omega_s^2-6\phi=(M_1\omega_s^2-6\phi)\mu_s \tag{2.3}$$

in which

$$\mu_s=\left[\frac{(\omega_s^2/\omega_2^2)-1}{(\omega_s^2/\omega_1^2)-1}\right]^{1/2}\ , \tag{2.4}$$

with $\omega_r=6\phi/M_r$ for $r=1,2$. In the above equations, $\mathbf{q}=(q_1,q_2,q_3)$ is the wave vector and the sum over $\mathbf{q}$ extends over the first Brillouin zone. In order to obtain the amplitude of the s-like localized mode we follow closely the approach introduced by Varma[12] to discuss the corresponding problem for defects in quantum crystals.

In the presence of a defect, the new normal modes for the perturbed lattice are introduced by the following canonical transformation:

$$\mathbf{u}(n)=\left[\frac{\hbar}{2}\right]^{1/2}\sum_s \mathbf{A}(s,n)\omega_s^{-1/2}(a_s+a_{-s}^\dagger)\ , \tag{2.5a}$$

$$\mathbf{p}(n)=i\left[\frac{\hbar}{2}\right]^{1/2}\sum_s \mathbf{A}^*(s,n)\omega_s^{1/2}M_n(a_s^\dagger-a_{-s})\ , \tag{2.5b}$$

where $a_s^\dagger$ (a_s) is the creation (annihilation) boson operator associated with the s mode, and the phonon mode amplitudes $\mathbf{A}(s,n)$ satisfy the following orthonormality conditions:

$$\sum_s M_nA_\alpha^*(s,n)A_\beta(s,m)=\delta_{\alpha\beta}\delta_{nm}\ , \tag{2.6a}$$

$$\sum_{n,\alpha} M_nA_\alpha^*(s,n)A_\alpha(s',n)=\delta_{ss'}\ . \tag{2.6b}$$

From Eqs. (2.5) we can note that in the harmonic approximation, with interaction between nearest neighbors, the amplitudes $\mathbf{A}(s,n)$ satisfy the time-independent equation of motion which describes the atomic vibration of our defective diatomic crystal model. In terms of the Green function these new $\mathbf{A}(s,n)$ can be written in the following form:

$$\mathbf{A}(s,l)=\sum_{n,m}(M_m\omega_s^2-6\phi)^{1/2}T(n,m)C(n,m)$$
$$\times g(\omega_s^2;l-n)\mathbf{A}(s,m)\ , \tag{2.7}$$

with

$$g(\omega_s^2;l-n)=(M_l\omega_s^2-6\phi)^{-1/2}G(\omega_s^2;l-n) \tag{2.8}$$

and

$$T(n,m)=\begin{cases}1, & \text{for } n+m=\text{odd}\\ \mu_s, & \text{for } n,m \text{ even}\\ 1/\mu_s, & \text{for } n,m \text{ odd}\end{cases} \tag{2.9}$$

the effect of the point imperfection is included in the C's which for an impurity atom located at the origin are given by[10]

$$C(0,0)=\epsilon M_1\omega^2+6\phi\xi\ ,$$
$$C(\pm100;\pm100)=-C(000;\pm100)=-C(\pm100;000)=C(0\pm10;0\pm10)=C(00\pm1;00\pm1)$$
$$=-C(000;0\pm10)=-C(0\pm10;000)=-C(000;00\pm1)=-C(00\pm1;000)=\xi\phi\ . \tag{2.10}$$

Now, we address ourselves to the task of obtaining the vibrational mode amplitudes associated with s-like localized modes at the impurity and at its neighboring sites, as well as deriving an asymptotic expression for $A_\alpha(s,n)$ with large $|\mathbf{R}_n|$. Using Eqs. (2.7)–(2.10) we can rewrite Eq. (2.7) as follows:

$$A_\alpha(s,n)=[\beta_sG(\omega_s^2;n)-(1-2\alpha_s)\xi\delta_{n,0}]A_\alpha(s,0)\ , \tag{2.11a}$$

for $n=$even

$$A_\alpha(s,n)=\mu_s^{-1}\beta_sG(\omega_s^2;n)A_\alpha(s,0)\ , \tag{2.11b}$$

for $n=$odd where

$$\beta_s=\mu_sM_1\omega_s^2(\epsilon+\xi)-2\mu_s\xi(M_1\omega_s^2-3\phi)\alpha_s\ , \tag{2.12}$$

$$\alpha_s=\frac{(\epsilon+\xi)M_1\omega_s^2G(\omega_s^2;1,0,0)}{1+2\xi(M_1\omega_s^2-3\phi)G(\omega_s^2;1,0,0)}\ . \tag{2.13}$$

In arriving at these equations, we have used the following identity satisfied by the Green functions:[11]

$$\phi\sum_\Delta G(\omega_s^2;l-\Delta)+6\phi v_s=\delta_{l,0}\ , \tag{2.14}$$

with

$$v_s=\left[\left[\frac{\omega_s^2}{\omega_1^2}-1\right]\left[\frac{\omega_s^2}{\omega_2^2}-1\right]\right]^{1/2} \tag{2.15}$$

and Δ means summation over all the nearest neighbors.

Using the orthonormality condition given in Eqs. (2.6), the coefficients $\mathbf{A}(s,n)$ at the impurity site are found to be

$$|\mathbf{A}(s,0)|^2=\left[\frac{1}{2}\left[\frac{\beta_s}{6\phi\mu_s}\right]^2(M_1\mu_s^2+M_2)\times F(v_s)-\epsilon M_1\right]^{-1}, \tag{2.16}$$

where

$$F(v_s)=\frac{1}{N}\sum_q (v_s+\gamma_q)^{-2}$$

and (2.17)

$$\gamma_q=\tfrac{1}{6}\sum_\Delta \cos\mathbf{q}\cdot\mathbf{R}_\Delta .$$

In the region of $v_s>1$ (localized s mode) an approximate expression for $F(v_s)$ has been evaluated by expanding the denominator in powers of γ_q/v_s.[13] To determine the asymptotic form of $A_\alpha(s,n)$, we note, from the form of the Green function in Eq. (2.2), that for large values of $|\mathbf{R}_n|$ the integrand oscillates rapidly as a function of $\mathbf{q}$ and so, throughout most of the Brillouin zone, the contributions from different q values tend to cancel. The exception to this comes from the region of q values for which the denominator is as small as possible. Therefore, following Callaway[14] and considering only s-like localized modes ($v_s>1$), we can express the Green function for sites away from the impurity in the form

$$G(\omega_s^2;n)=\frac{-a}{4\pi\phi}h(\rho_s)\frac{e^{-\rho_s R_n}}{R_n},\quad R_n\gg 0 \tag{2.18}$$

where a denotes the lattice constant and ρ_s is determined by the equation

$$v_s=\gamma_{q=i\rho_s},\quad v_s>1 . \tag{2.19}$$

With the aim of studying explicitly the dependence of some quantities on the energy v_s, we will write the inverse range parameter ρ_s as an approximate solution of Eq. (2.19), i.e.,

$$\rho_s=a^{-1}[6(v_s-1)]^{1/2} , \tag{2.20}$$

which is valid for $qa\ll 1$. In this limit the function $h(\rho_s)$ is of the order of one.[14] As we expected, in the case of states with frequencies above the band of the ideal lattice, the vibrational mode amplitudes of the atoms [Eqs. (2.11)] decrease exponentially with increasing distance from the defect.

III. PARAMETRIC EXCITATION OF PHONONS VIA LOCALIZED MODES

Sparks and Chow[15] were the first to study, from a theoretical point of view, the phonon instabilities in ionic crystals such as NaCl caused by the anharmonic interaction. In particular, they considered the effect of parametric instabilities of phonons on the optical absorption which is of considerable interest in nonlinear optical effects. From the theoretical analysis, it has been possible to obtain the threshold for the parametric instability as a function of the amplitude and lifetime of the fundamental phonons. In this section we present the derivation of the anharmonic interaction between a localized s mode and two-phonon waves of the perfect lattice, which will be used to deduce the critical number of pumped impurity modes at the threshold of the nonlinear process.

The whole Hamiltonian of the system under study is now assumed to include, in addition to the harmonic part, the anharmonic interaction

$$H_I=\tfrac{1}{12}\sum_{\substack{n,m\\ \alpha,\beta,\gamma}}\phi_{\alpha\beta\gamma}(n,m)u_\alpha(n,m)u_\beta(n,m)u_\gamma(n,m) , \tag{3.1}$$

where $\mathbf{u}(n,m)=\mathbf{u}(n)-\mathbf{u}(m)$ and $\phi_{\alpha\beta\gamma}(n,m)$ is the anharmonic coupling which for any central force potential is given explicitly by

$$\phi_{\alpha\beta\gamma}(n,m)=\frac{\partial^3}{\partial\alpha\partial\beta\partial\gamma}\phi(r)\Big|_{\mathbf{r}=\mathbf{R}(n,m)}$$
$$=\left[\frac{\alpha\beta\gamma}{r^3}\left[\phi'''(r)-\frac{3}{r}\phi''(r)+\frac{3}{r^2}\phi'(r)\right]+\left[\delta_{\alpha\beta}\frac{\gamma}{r^2}+\delta_{\beta\gamma}\frac{\alpha}{r^2}+\delta_{\gamma\alpha}\frac{\beta}{r^2}\right]\left[\phi''(r)-\frac{1}{r}\phi'(r)\right]\right]_{\mathbf{r}=\mathbf{R}(n,m)} . \tag{3.2}$$

Leibfried[16] has pointed out, in connection with a discussion of a cubic anharmonic term for central force models, that

$$|a\phi'''(a)/\phi''(a)|\sim 10 ; \tag{3.3}$$

thus, it is a good approximation to retain only the highest-order derivative in Eq. (3.2), i.e.,

$$\phi_{\alpha\beta\gamma}(n,m)=\frac{R_\alpha(n,m)R_\beta(n,m)R_\gamma(n,m)}{|\mathbf{R}(n,m)|^3}\times\phi'''(|\mathbf{R}(n,m)|) , \tag{3.4}$$

where $\mathbf{R}(n,m)=\mathbf{R}_n-\mathbf{R}_m$ is the relative distance between ions at the equilibrium position and $\phi'''(r)$ represents the third derivative of the interaction potential between two atoms separated by a distance r. The Hamiltonian H_I can now be expressed in terms of the creation and annihilation operators through the canonical transformation (2.5) which can conveniently be rewritten in such a way as to separate out the elementary excitations of the systems

$$\mathbf{u}(n)=\sum_Q\left[\frac{\hbar}{\omega_Q}\right]^{1/2}\mathbf{A}(Q,n)(a_Q+a^\dagger_{-Q})+\sum_s\left[\frac{\hbar}{\omega_s}\right]^{1/2}\mathbf{A}(s,n)(a_s+a^\dagger_{-s}) . \tag{3.5}$$

Here $\mathbf{A}(Q,n)$ is the phonon wave amplitude for the pure crystal which is given by

$$\mathbf{A}(Q,n)=\frac{1}{(NM_n)^{1/2}}\epsilon_n(Q)e^{-i\mathbf{q}\cdot\mathbf{R}_n} \tag{3.6}$$

and $Q=(\mathbf{q},j)$ specifies the phonon mode with wave vector $\mathbf{q}$ on branch j of the host lattice, with $-Q=(-\mathbf{q},j)$. In the following, for the sake of simplicity, we assume a substitutional isotopic impurity atom, i.e., $\xi=0$, and retaining only terms which can destroy the localized modes, we obtain

$$H_I=\sum_{s,Q_1,Q_2}[V(s,Q_1,Q_2)a_s a^{\dagger}_{-Q_1}a^{\dagger}_{-Q_2}+\text{H.c.}]\,, \tag{3.7}$$

where

$$V(s,Q_1,Q_2)=\frac{\phi'''(a)}{2}\sigma(s,Q_1,Q_2)\times\sum_{m=1}^{6}\prod_{i=1}^{2}U_m(Q_i)\hat{\mathbf{R}}_m\cdot\mathbf{A}(0)\times\left[\frac{\hbar}{2N\omega_{Q_i}M_1}\right]^{1/2}, \tag{3.8}$$

with

$$\sigma(s,Q_1,Q_2)=\frac{\beta_s h(\rho_s)(1+\mu_s)}{\phi\mu_s}\left[\frac{\hbar}{\omega_s}\right]^{1/2}\times\frac{a^{-2}}{\rho_s^2+|\mathbf{q}_1+\mathbf{q}_2|^2}\,, \tag{3.9}$$

$$U_m(Q_i)-\hat{\mathbf{R}}_m\cdot\left[\epsilon_1(Q_i)-\left[\frac{M_1}{M_2}\right]^{1/2}\times\epsilon_2(Q_i)e^{-i\mathbf{q}_i\cdot\mathbf{R}_m}\right]. \tag{3.10}$$

In Eq. (3.7) the position of nearest-neighbor atoms measured from the lighter ion ($M_1<M_2$) are $\mathbf{R}_m$, $\hat{\mathbf{R}}_m$ is the unit vector in the same direction, and $\epsilon_n(Q)$ represents the polarization vector of the nth ion. In writing the above results we have used the asymptotic form of the Green function given in Eq. (2.18) which is a good approximation for treating the atomic motion around the impurity.

Equation (3.7) represents the decay of a localized phonon in a pair of phonons of the host lattice and, as expected, the wave vector is not necessarily conserved in this process. However, it can be seen that the maximum amplitude in $V(s,Q_1,Q_2)$ occurs for $q_1=-q_2$, meaning that this is the most probable process and therefore it produces the lowest threshold for the parametric instability of phonons. This threshold condition can be obtained from the instability criterion[15] which can be applied to any boson-boson process, i.e., instability occurs when the number of quanta in the q mode required to maintain equilibrium becomes very large. In what follows we shall deduce the critical number of localized modes, N_c, pumped by a strong electromagnetic field at the parametric instability threshold.

First consider the power flow from the localized s mode to the potentially unstable host phonon waves. Using the standard first-order perturbation theory, the rate of change of the number of localized modes N_s resulting from the coupling to a single pair of phonons Q_1 and Q_2 is given by

$$\frac{dN_s}{dt}=\frac{2\pi}{\hbar^2}\{|\langle\dot{N}_s+1,N_{Q_1}-1,N_{Q_2}-1|H_I|N_s,N_{Q_1},N_{Q_2}\rangle|^2-|\langle N_s-1,N_{Q_1}+1,N_{Q_2}+1|H_I|N_s,N_{Q_1},N_{Q_2}\rangle|^2\}\delta(\omega_s-\omega_{Q_1}-\omega_{Q_2}) \tag{3.11}$$

and the rate of change of N_Q from the coupling to the localized mode is

$$\hbar\omega_{Q_1}\left[\frac{dN_{Q_1}}{dt}\right]+\hbar\omega_{Q_2}\left[\frac{dN_{Q_2}}{dt}\right]+\hbar\omega_s\left[\frac{dN_s}{dt}\right]=0\,. \tag{3.12}$$

Now, in equilibrium, the net rate of change of N_Q must be zero. Thus, the rate of increase of the host phonon system is added to the rate of decrease by relaxation

$$\left[\frac{dN_Q}{dt}\right]_{\text{relax}}=-\eta_Q(N_Q-\tilde{N}_Q) \tag{3.13}$$

and the result is set equal to zero to obtain two equations

$$CN_0=\eta_{Q_1}(N_{Q_1}-\tilde{N}_{Q_1})\,, \tag{3.14a}$$

$$CN_0=\eta_{Q_2}(N_{Q_2}-\tilde{N}_{Q_2})\,, \tag{3.14b}$$

where

$$N_0=N_s+N_sN_{Q_1}+N_sN_{Q_2}-N_{Q_1}N_{Q_2}\,,\qquad C=2\pi|2\phi(s,q,j_1,J_2)|^2\delta(\omega_s-\omega_{Q_1}-\omega_{Q_2})\,, \tag{3.15}$$

and η_Q is the relaxation frequency of an output phonon produced by the decaying process of the impurity mode, and the coupling coefficient is given by

$$\phi(s,qj_1,j_2)=\hbar^{-1}V(s,qj_1,-qj_2)\,.$$

Here we have considered only the processes in which $q_1=-q_2=q$ since it produces the lowest threshold. $\tilde{N}_Q$ is the Bose-Einstein distribution function. The coupled algebraic equations (3.14) can easily be solved by substitution, which gives

$$N_{Q_1}=A\{\text{sgn}A[1+(\eta_{Q_2}\tilde{N}_{Q_2}-\eta_{Q_1}\tilde{N}_{Q_1}+\eta_{Q_2}N_s+\eta_{Q_1}\eta_{Q_2}\tilde{N}_{Q_1}C^{-1})/\eta_{Q_1}A^2]^{1/2}-1\}\,,$$

$$N_{Q_2}=\frac{\eta_{Q_1}}{\eta_{Q_2}}(N_{Q_1}-\tilde{N}_{Q_1})+\tilde{N}_{Q_2}\ , \tag{3.16}$$

where

$$A=[-N_s(\eta_{Q_1}+\eta_{Q_2})+\eta_{Q_2}\tilde{N}_{Q_2}-\eta_{Q_1}\tilde{N}_{Q_1} +\eta_{Q_1}\eta_{Q_2}C^{-1}]/2\eta_{Q_1} \tag{3.17}$$

and sgn$x$ represents the sign function of x. Equation (3.16) has the following limiting values.

(i) If $\eta_{Q_1}=\eta_{Q_2}=\eta_Q$, then

$$N_Q\simeq\tilde{N}_Q,\ \text{for } N_s\ll N_c \tag{3.18a}$$

$$N_Q=[N_c(2\tilde{N}_Q+1)]^{1/2},\ \text{for } N_s=N_c \tag{3.18b}$$

$$N_Q=2(N_s-N_c),\ \text{for } N_s\gg N_c \tag{3.18c}$$

and (ii) if $\eta_{Q_1}\gg\eta_{Q_2}$, then

$$N_{Q_1}\simeq\tilde{N}_{Q_1},\ \text{for } N_s\ll N_c \tag{3.19a}$$

$$N_{Q_1}=\left[N_c\left[\frac{\eta_{Q_1}}{\eta_{Q_2}}+\tilde{N}_{Q_1}\right]\right]^{1/2}+\frac{\tilde{N}_{Q_1}}{2}\ , \tag{3.19b}$$

for $N_s=N_c$

$$N_{Q_1}=N_s+\tilde{N}_{Q_1}\frac{N_c}{N_s}+\frac{\eta_{Q_1}}{\eta_{Q_2}},\ \text{for } N_s\gg N_c \tag{3.19c}$$

with

$$N_c=\eta_{Q_1}\eta_{Q_2}/(\eta_{Q_1}+\eta_{Q_2})C\ .$$

The approximations (i) and (ii) represent a process in which the localized mode pumped by the external electromagnetic radiation decays into two phonons of equal frequency and unequal frequency, respectively. From Eqs. (3.18) and (3.19) we shall say that the two-phonon process has been driven unstable if $N_s>N_c$, and the threshold of the instability is defined by $N_s=N_c$. This threshold value N_c in Eqs. (3.19) is the same for both modes since N_{Q_1} and N_{Q_2} are linearly related, as seen in Eq. (3.16b). This instability criterion can be seen clearly by linearizing the equation of motion. In the present case, linearizing Eq. (3.14) by neglecting the term $N_{Q_1}N_{Q_2}$ gives that N_Q is proportional to $(N_c-N_s)^{-1}$ which displays instability at $N_s=N_c$ vividly since $N_Q\to\infty$ there. Using the results of these equations, we find that the critical number of the pumped localized modes N_s^{crit} at the resonance is given by

$$N_s^{\text{crit}}=\frac{\eta_Q^2}{32\,|\,\phi(s,Q)\,|^2},\ \text{for } \eta_{Q_1}=\eta_{Q_2}=\eta_Q \tag{3.20a}$$

and

$$N_s^{\text{crit}}=\frac{\eta_{Q_1}\eta_{Q_2}}{64\,|\,\phi(s,q,j_1,j_2)\,|^2},\ \text{for } \eta_{Q_1}\gg\eta_{Q_2}\ . \tag{3.20b}$$

In arriving at these equations, we have assumed a normalized line-shape factor for the δ function evaluated at resonance,[15] i.e.,

$$\delta(\omega_s-\omega_{Q_1}-\omega_{Q_2})=\frac{2}{\pi}\frac{\omega_s(\omega_{Q_1}+\omega_{Q_2})(\eta_{Q_1}+\eta_{Q_2})}{[\omega^2-(\omega_{Q_1}+\omega_{Q_2})^2]^2+(\omega_{Q_1}+\omega_{Q_2})^2(\eta_{Q_1}+\eta_{Q_2})^2}\simeq\frac{2/\pi}{\eta_{Q_1}+\eta_{Q_2}}\ . \tag{3.21}$$

In order to relate the threshold condition $N_s=N_c$ to the experimentally controllable quantity, which in this case is the intensity I of the external incident radiation, the relation between I and N_s is first derived as follows: If the intensity just at the face is I_0, the intensity at a distance x into the sample with thickness $\gg 1/\beta$ is $I=I_0e^{-\beta x}\simeq I_0-\beta x I_0$, where β is the absorption coefficient and the approximate equality holds for $\beta x\ll 1$. The rate of energy absorption in the layer of volume Ax is $-A(I-I_0)=Ax\beta I$, where $I_0\simeq I$. Equating this rate of energy absorbed by the localized mode to the rate of loss by relaxation

$$\hbar\omega\eta_s(N_s-\tilde{N}_s)Ax/V\simeq\hbar\omega N_s Ax/V\ ,$$

where V is the volume of the sample, gives

$$I=\hbar\omega N_s\eta_s/V\beta(\omega) \tag{3.22}$$

therefore, with $N_s=N_c$ in Eq. (3.22) we get the critical intensity I_c at resonance, i.e., $\omega=\omega_s$. Here $\beta(\omega)$ is the impurity induced infrared absorption coefficient of alkali halide crystal which, in the limit of small-impurity concentration is given by[17]

$$\beta(\omega)=\frac{4\pi e^2N_i}{n_rcV}\frac{\omega^5K(\omega)^2\mu_sG_0''(\omega)}{[\text{Re}D(\omega^2)]^2+[\text{Im}D(\omega^2)]^2},\ \omega\neq\omega_m \tag{3.23}$$

where c is the light velocity, n_r is the index of refraction of the crystal, N_i is the number of impurity atoms, ω_m is the frequency of the top of the optical band. Re$D(\omega^2)$ and Im$D(\omega^2)$ are abbreviations of

$$\text{Re}\{\lim_{\delta\to 0+}D[(\omega+i\delta)^2]\}$$

and

$$\text{Im}\{\lim_{\delta\to 0+}D[(\omega+i\delta)^2]\}\ ,$$

respectively [$D(\omega^2)$ is defined in Eq. (2.1)]. $G_0''(\omega)$ is the imaginary part of the Green function evaluated at the origin and $K(\omega)$ is given by

$$K(\omega)=\frac{e^*(1-\omega)}{\omega_1^2}+\frac{\xi+\epsilon+(M_1/M_2)\xi(1-\epsilon)}{\omega^2-\omega_m^2}\ , \tag{3.24}$$

where $e^*=[(e'/e)-1]$ is the parameter characterizing the change of the impurity charge e', as compared with the charge of the host lattice e.

Note that, after experimental determination of the physical parameter I_c, it is possible to obtain information about the coupling coefficients between localized s modes and phonon waves, as well as measurements of the relaxation rates of impurities.

IV. SUMMARY

With the aid of specific crystal and impurity models, a theory of nonlinear phonon generation via localized modes of a diatomic cubic crystal lattice has been developed. These processes are characterized by a growth of the number of phonons in the unstable mode due to the decay of localized modes via anharmonic interaction. In this paper an analytical expression for the critical intensity in terms of the relaxation frequency of the s mode and the absorption coefficient has been obtained. Comparing the result of I_c with experiment, we can, in principle, determine η_s and therefore the coupling coefficients between the impurity atom and its nearest neighbors.

To make an order of magnitude estimate of the power of an infrared laser source necessary to attain the critical intensity of photon given in Eq. (3.9), let us consider an estimate of the matrix element $V(s,Q_1,Q_2)$ which is related to the relaxation frequency η_s of the localized mode in the following form: Using the interaction term of Eq. (3.4) and from the standard time-dependent perturbation theory result for the transition probability between the localized s mode and the phonon states of the host crystal, η_s is given by

$$\eta_s=\frac{4\pi}{\hbar}\sum_{Q_1,Q_2}|V(s,Q_1Q_2)|^2(\tilde{N}_{Q_1}+\tilde{N}_{Q_2}+1)\times\delta(\hbar\omega_s-\hbar\omega_{Q_1}-\hbar\omega_{Q_2}), \tag{4.1}$$

where

$$\tilde{N}_Q=|\exp(\hbar\omega_Q/k_BT)-1|^{-1}$$

is the thermal equilibrium occupation number and

$$\tilde{N}_{Q_1}+\tilde{N}_{Q_2}+1=\frac{1}{2}\frac{\sinh(\hbar\omega_s/2k_BT)}{\sinh(\hbar\omega_{Q_1}/2k_BT)\sinh(\hbar\omega_{Q_2}/2k_BT)}. \tag{4.2}$$

Now, summing Eq. (4.1) over $q=-q_1=q_2$ (the most probable process) and considering the decay process into phonons on different branches,[18] for example, one acoustic and one optical branch, we obtain

$$|\phi(s,q,J_1,J_2)|^2V=h^{-2}|V(s,q,J_1,J_2)|^2V=\frac{\eta_s\pi t}{2\left[\frac{\omega_s-\omega_0}{t}\right]^2(\tilde{N}_0+\tilde{N}_Q+1)}. \tag{4.3}$$

The integral above was calculated using a linear dispersion relation for acoustic phonons ($\omega_Q=tq$) and the Einstein frequency for the optical phonons ($\omega_Q=\omega_0$). To make an order of magnitude calculation, let us take the following typical parameters for NaCl at room temperature: $\eta_{\omega=\omega_0}=2.10^{12}$ sec^{-1}, $\eta_{\omega=tq}=2.10^{11}$ sec^{-1}, $\omega_0=3.10^{13}$ sec^{-1}, $t=10^5$ cm/sec, $\eta_s=7.6\times10^{12}$ sec^{-1}, $\tilde{N}_0+\tilde{N}_Q+1=5.6$, and $\beta(\omega)\simeq1$ cm^{-1}.[19] According to Eqs. (3.20), (3.22), and (4.3) on resonance $I_c=10^{13}$ W/cm^2. This is a large value of I_c, and in experiments to look for the instability, a small value of I_c is desirable. Thus, smaller values of η_s and η_Q are needed. For small q acoustical modes on the lowest frequency branch at low temperature, η_Q is believed to be small.[15] A conservative approximation to the lower limit of η_Q is 10^9 sec^{-1}. At low temperature, η_s will also be smaller, say, by a factor of 5 smaller than at room temperature. Therefore, I_c will be of the order of 10^8 W/cm^2 which can be obtained using pulse techniques.

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