

## Coherent production of phonons from time-dependent strains

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A simple model of a classical oscillator, with time-dependent frequency, is applied to the normal modes of condensed systems. Taking advantage of the analogy with the stationary Schrödinger equation in one dimension, the existence of special modes (*s* modes), with *exponentially increasing* amplitude, is proven, both for disordered and for periodic time-dependent strains. Anharmonic effects are then accounted for in terms of absorption processes for the excess phonons (*s* phonons), coherently produced by the strain. A nonmetallic continuum lattice is used as a reference system for applications. Possible macroscopic effects, such as the resonant activation of optical modes and the production of time-controlled shock waves, are outlined. [S0163-1829(97)02826-9]

### I. INTRODUCTION

The recent debate about cold fusion<sup>1-4</sup> raised the question whether it is at least conceivable for a metallic system to transfer such a large energy to light ions, as to enhance the rate of any nuclear reaction up to significant values. This question has stimulated a number of unusual insights, about the way in which the elastic energy can be accumulated and released, under certain critical conditions. In particular, Cas-sandro, Gallavotti, and Jona-Lasinio<sup>5</sup> and Maiani, Parisi, and Pietronero,<sup>6</sup> put forth an explanation of the experiment of De Ninno *et al.*,<sup>3</sup> based on a local structural phase transition, with a sudden release of elastic energy, in the form of shock waves. The energy of the shock wave should be initially stored at the surface of small hydrogen-rich dots. Statistical arguments are then used by Tabet and Tenebaum,<sup>7</sup> to justify the crucial point of the question, that is, the *coherence* of the energy release. The shock wave in Refs. 5-7 can indeed be regarded as a sort of phononic laser, whose quanta are spherical phonons imploding from the border of a small critical region to its center. Apart from any possible application to the cold fusion, the preceding arguments do actually open a stimulating field of investigation, about the possibility that *coherently produced* phonons do accumulate (and release) large amounts of energy, well above the "natural" thermal values characteristic of each phonon. The present paper aims to approach this problem from a general viewpoint.

In Sec. II the "storage" of the elastic energy is approached, as a preliminary step to the coherent production of phonons. The basic mechanism of energy storage is generated by a very simple mathematical model, that is, a classical oscillator whose elastic constant (or proper frequency) is affected by time-dependent fluctuations. It will be shown that, under very general conditions, the energy of the oscillator may increase *exponentially* in time, at the expense of the external source producing the frequency fluctuations, in the absence of dissipative processes. In Sec. III, we discuss the possible physical realization of the source, while in Secs. IV and V the anharmonic effects are accounted for in terms of phonon-phonon scattering processes. In Sec. VI, the coherent

production of phonons is studied for an ideal continuum lattice model (CLM). Indications for macroscopic effects of the coherent production of phonons are given.

### II. ENERGY STORAGE

The *classical* equation of an oscillator with time-dependent frequency reads

$$-\frac{d^2Q}{dt^2} = [\Omega_0^2 + \delta(t)]Q, \quad (1)$$

where  $Q$  is any generalized coordinate of the system, and  $\delta(t)$  is a suitable function of time. We will discuss in Sec. III some possible physical realizations of Eq. (1). For the moment, we simply study the nature of the solutions, according to the behavior of  $\delta(t)$ . First of all, we justify the use of the classical approximation (for an outline of the quantum case, see the end of the present section) by assuming that the system is in thermal contact with a bath at temperature  $T$  such that  $\kappa T \gg \hbar(\Omega_0^2 + \delta_M)^{1/2}$ ,  $\delta_M$  being the maximum value attained by  $|\delta(t)|$ . Though *classical* in nature, Eq. (1) can be formally mapped into a *quantum* problem, that is, the stationary Schrödinger equation in one dimension, provided

$$\begin{aligned} t \text{ (time)} &\rightarrow x \text{ (spatial coordinate),} \\ Q(t) &\rightarrow \psi_E(x) \text{ (energy eigenfunction),} \\ \Omega_0^2 &\rightarrow E \text{ (energy eigenvalue)} \times 2m/\hbar^2, \\ -\delta(t) &\rightarrow V(x) \text{ (potential energy)} \times 2m/\hbar^2. \end{aligned} \quad (2)$$

The mapping Eq. (2) is very convenient for the present aims, since the nature of the solutions of the corresponding Schrödinger equation are well known, if  $V(x)$  fluctuates without limit in a *finite* range of values, for  $x \rightarrow \pm\infty$ . If the fluctuations are disordered (in any sense), for any value of  $E$  belonging to a set  $\{E_{\text{loc}}\}$  of "allowed" values, the eigensolutions  $\psi_E(x)$  are *localized*, that is, their envelope is exponentially decreasing for  $x \rightarrow \pm\infty$  (Anderson localization in one dimension<sup>8</sup>). Otherwise, any solution of the

Schrödinger equation is *exponentially increasing* in  $|x|$ , apart from possible *isolated* values  $\{E_{\text{ext}}\}$  corresponding to *extended* solutions, that fluctuate periodically in a limited range of values for  $x \rightarrow \pm\infty$ .<sup>9</sup> Apart from the isolated point spectrum  $\{E_{\text{ext}}\}$ , the general solution of the Schrödinger equation in case a always include *exponentially diverging* components, whose elimination on both sides of the  $x$  axis, is possible for  $E \in \{E_{\text{locf}}\}$  only, and for very special choices of the initial conditions. Coming back to Eq. (1), by means of Eq. (2), we can conclude as follows:

(a) If  $\delta(t)$  fluctuates *disorderly* in time, within a limited range of values, the envelope of  $Q(t)$  increases exponentially in time, for almost all initial conditions and unperturbed frequencies.

The other case of interest for the present aims is a periodic potential  $V(x) = V(x+a)$ , in the “equivalent” Schrödinger equation. If the eigenvalues  $E$  belong to special intervals, denoted as “allowed bands,” there are acceptable (in the quantum sense) solutions  $\psi_E(x)$ , expressed as a linear combination of two Bloch functions, whose square modulus have the same periodicity  $a$  as the potential  $V(x)$ . If, instead,  $E$  belongs to the complementary intervals, denoted as “forbidden bands” (or gaps), the general solution is a linear combination of two functions, whose envelopes *diverge exponentially* for  $x \rightarrow +\infty$  or  $x \rightarrow -\infty$ . Allowed bands and gaps are alternate on the  $E$  axis, without upper limit, and can be thereby labeled by an index  $j = 1, 2, \dots$ . In conclusion, according to the mapping (2):

(b) If  $\delta(t)$  is a *periodic* function of time of period  $\tau$ , the nature of the solutions of Eq. (1) is twofold: if  $\Omega_0^2$  falls into an “allowed band” of the periodical “potential”  $-\delta(t)$ , the coordinate  $Q(t)$  is a periodic function of time, of period  $2\pi/\Omega_0$  (the unperturbed solution), modulated by another periodic function of period  $\tau$ . Instead, if  $\Omega_0^2$  falls into a “gap” of the periodic potential  $-\delta(t)$ ,  $Q(t)$  is split into two oscillating components, with unperturbed period  $2\pi/\Omega_0$ , one of which is modulated by an *exponentially increasing* function of time, while the other is damped by an exponentially decreasing function. At long times, it is clear that the former will always dominate, except for very special initial conditions, having zero measure in phase space, for which the exponentially diverging component has rigorously zero weight at  $t=0$  (the initial time at which the time-dependent stress is applied).

In case (b), it is possible to study the exponentially increasing solutions on the assumption that  $\delta_M = \max\{|\delta(t)|\} \ll \Omega_0^2$  (small fluctuations), and for the model sketched in Fig. 1:  $\delta(t)$  is therein depicted as a piecewise function (for  $t > 0$ ), such that “rectangular wells” of depth  $\delta_M$  and duration  $\Delta\tau$ , appear periodically, with period  $\tau (> \Delta\tau)$ . In this case, the quantum problem equivalent to Eq. (1) [according to the mapping Eq. (2)], becomes a standard exercise for undergraduate students.<sup>10</sup> First, Eq. (1) can be put in a dimensionless form:

$$\stackrel{\text{def}}{s} = \Omega_0 t; \quad \stackrel{\text{def}}{\Phi}(s) = Q(s/\Omega_0) \Rightarrow -\frac{d^2\Phi}{ds^2} = \left[ 1 + \frac{\delta(s/\Omega_0)}{\Omega_0^2} \right] \Phi. \quad (3)$$

Now the question is whether value 1 does or does not belong to one of the allowed bands generated by a distribution of

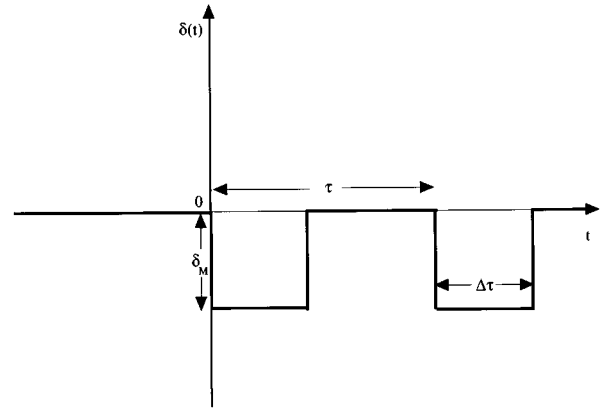


FIG. 1. A “potential-barrier” model for the fluctuations in time of the square proper frequency of an oscillator. The perturbation starts at  $t=0$ .

rectangular barriers (wells), of height (depth)  $\xi = -\delta_M/\Omega_0^2$ , width  $\Delta s = \Omega_0 \Delta\tau$ , and periodically distributed on the  $s$  axis, with period  $s_0 = \Omega_0 \tau$ . Setting  $s' = s_0 - \Delta s$ , it can be easily shown that the solution of the problem stated above is given by the quantity

$$R = \cos(s') \cos(\Delta s \sqrt{1 - \xi}) - \frac{1 - \xi/2}{\sqrt{1 - \xi}} \sin(s') \sin(\Delta s \sqrt{1 - \xi}). \quad (4)$$

Value 1 does or does not belong to an allowed band according to whether  $R^2 \leq 1$  or  $R^2 > 1$  respectively. In the latter case, the rate of the exponential increase in  $s$  of  $\Phi$  [Eq. (3)] is  $s_0^{-1} \ln[|R| + \sqrt{R^2 - 1}]$ .<sup>11</sup> Since  $\xi$  is assumed small, we can study Eq. (4) to the first significant order (i.e.,  $\xi^2$ ). It follows that the condition  $R^2 > 1$  is realized when the quantity  $s_0 - \Delta s \xi/2$  falls in small intervals of width  $|\xi \sin(s')|$ , around the special values  $j\pi$  ( $j = 0, \pm 1, \pm 2, \dots$ ). The resulting rate of increase (in  $s$ ) is  $s_0^{-1} |\xi \sin(\Delta s)|/2$ , to the lowest order in  $\xi$ . Recalling the definitions in Eq. (3), and coming back to the  $t$  scale, we can conclude that the oscillator Eq. (1), in the model fluctuation Fig. 1, becomes a *special* mode, adsorbing energy exponentially in time, if

$$\Omega_0 \tau + \frac{\Delta\tau \delta_M}{2\Omega_0} = \pi j \quad (j = 0, 1, 2, \dots) \quad (\text{resonance condition}), \quad (5a)$$

for which the rate of exponential increase (in time) of the  $Q$  coordinate becomes

$$\omega_s = \frac{\delta_M}{2\Omega_0^2 \tau} |\sin(\Omega_0 \Delta\tau)|. \quad (5b)$$

Equation (5a) is not strictly a “resonance” condition, since there is a frequency band of width  $|\xi \sin(\Omega_0(\tau - \Delta\tau))| \tau^{-1}$ , around each value in the right-hand side (rhs) of Eq. (5a), for which an exponential increase applies as well. The value (5b) is, indeed, the *maximum* rate within the band. Since  $\xi = -\delta_M/\Omega_0^2$  is our smallness parameter, in the present case it is justified to neglect any bandwidth effect (these will be reconsidered in Sec. V). From now on, we will call “ $s$  modes” all the special modes, whose amplitude’s envelop increases ex-

ponentially in time under the action of Eq. (1). However, we anticipate that dissipation effects will, in general, suppress the exponential rate of increase, as discussed in Secs. IV and V.

At first sight, it might appear far more convenient to refer to *disordered* fluctuations [case (a)], than to *periodical* fluctuations [case (b)], in view of an exponential energy storage in the oscillator Eq. (1). In fact, case (a) gives rise to the required effect for *all* values of  $\Omega_0\tau$ , except at most for a zero measure set. Instead, case (b) requires a quiresonance condition around *isolated* values of  $\Omega_0\tau$  (for small periodical fluctuations), and looks thereby much more selective. In practice, however, the situation is not so different. If we assume *small disordered* fluctuations in case (a) too, it is possible to see that the disorder is a second-order effect ( $\propto \xi^2$ ), with respect to a suitable *periodic* potential, denoted as a coherent potential.<sup>8</sup> The periodic problem (coherent potential approximation), yields the same results as in case (b), that is, a resonance condition on  $\Omega_0\tau$ , giving rise to a rate of exponential increase  $\omega_s \propto \xi$  [Eq. (6c)]. The reason for the general exponential increase at *all*  $\Omega_0$  is that in the present *one-dimensional* case, the disorder itself, no matter how small, is shown to produce a finite rate of exponential increase  $\omega_{\text{dis}} \propto \xi^2$ .<sup>8</sup> However, it follows that  $\omega_{\text{dis}} \propto \xi^2$ , while  $\omega_s \propto \xi$ . Thus in case (a), we expect that for  $\Omega_0\tau$  fulfilling the condition (5a), there is a rate of exponential increase  $\omega_s \propto \xi$ , *much larger* than the rate  $\omega_{\text{dis}} \propto \xi^2$ , corresponding to the  $\Omega_0\tau$ 's not fulfilling the same condition. This will have relevant consequences when the dissipation effects are accounted for.

As a concluding remark on the elementary mechanism of energy storage, we comment about the validity of the *classical* approximation underlying Eq. (1). Does the exponential increase of the energy hold true for a *quantum* oscillator too? The question can be approached from the well-known result that the quantum mean energy of a harmonic oscillator *cannot* be smaller than the corresponding classical mean energy. The exponential increase of the latter is thereby a sufficient condition for the former to increase *at least* with the same rate. However, the quantitative aspects of the quantum oscillator's problem with fluctuating frequency are far from trivial, and require a separate analysis, which will appear in a forthcoming paper.

### III. PHYSICAL ORIGIN OF THE FREQUENCY FLUCTUATIONS

Before discussing the applications of the preceding arguments to microscopic scales, it is worth stressing that Eq. (1) describes, in the small oscillation limit, the dynamics of a child sitting on a swing.<sup>12</sup> In fact, the child's movements can actually change the swing frequency in time, by changing the inertial moment. In agreement with cases (a) and (b), discussed in Sec. II, it is a matter of common experience that moving disorderly [case (a)] is not a good strategy for increasing the oscillation amplitude. The resulting exponential rate is indeed small, and may be suppressed by all dissipative effects influencing a real swing. Instead, there are optimal *periodic* movements [case (b)], corresponding to the resonance condition (5a), which yield an effective rate of in-

crease, and quickly drive the swing out of the small oscillation regime.

Coming back to microscopic scales, Eq. (1) can be applied to *normal* elastic modes in condensed systems, provided the applied stress field does produce an *adiabatic* time-dependent strain, such that, at each instant, the normal elastic modes themselves can be defined, even in the strained system. For this to occur, the rate of change of the strain must be small compared to the proper frequency of any mode. For a periodic strain of period  $\tau$ , the condition above reads  $\tau \gg \max\{\Omega_0^{-1}\}, \{\Omega_0\}$  being the set of unperturbed proper frequencies. If the strains are nonadiabatic, it is impossible to separate the elastic part of the Hamiltonian from the interaction part coupling the atomic coordinates to the stress field. In this case, *viscoelastic* effects, which we neglect here at all, should be accounted for. In conclusion, an adiabatic strain is one producing *anharmonic* effects only, as we will assume in what follows.

The proper frequency of a normal mode, in a crystalline material, has the general form  $\Omega(\mathbf{k}) = \Omega(\{\mathbf{a}_\alpha\}; \{\mathbf{ka}_\alpha\})$ , where the  $\mathbf{a}_\alpha$ 's are the elementary lattice vectors ( $\alpha = 1, 2, 3$ ). For an adiabatic strain preserving the crystal symmetry, such that

$$\mathbf{a}_\alpha(t) = \mathbf{a}_{0\alpha} + \Delta\mathbf{a}_\alpha(t), \quad (6a)$$

it is immediately seen that the scalar products  $\mathbf{ka}_\alpha$ 's are invariant, since the wave vectors  $\mathbf{k}$ , determined (for example) by periodic boundary conditions, do change with time as well, in such a way that  $\mathbf{ka}_\alpha = 2\pi(n/N)$ ,  $n$  being an integer, and  $N$  being the number of unit cells in the lattice. As stressed above, changes in the proper frequency  $\Omega(\{\mathbf{a}_\alpha\}; \{\mathbf{ka}_\alpha\})$  can only come from the *anharmonic* effects accounted for by the explicit dependence on the  $\mathbf{a}_\alpha$ 's themselves. For example, Grüneisen's theory takes the density  $\rho$  as the most important quantity related to the  $\mathbf{a}_\alpha$ 's, and assumes that

$$\Omega(\{\mathbf{a}_\alpha\}; \{\mathbf{ka}_\alpha\}) = \Omega(\rho, \{\mathbf{ka}_\alpha\}) = \left(\frac{\rho}{\rho_0}\right)^\gamma \Omega_0(\rho_0, \{\mathbf{ka}_\alpha\}), \quad (6b)$$

where  $\rho_0$  is a reference density and  $\gamma (\cong 2)$  is the Grüneisen parameter. Within the limits of validity of this approximation, one may take  $\rho_0$  as the unperturbed density, and introduce a time-dependent uniform pressure, such that  $\rho(t) = \rho_0 + \Delta\rho(t)$ . From Eq. (6b) it follows that

$$\Omega(t, \mathbf{k}) \cong \Omega_0(\mathbf{k}) \left[ 1 + \gamma \frac{\Delta\rho(t)}{\rho_0} \right], \quad (6c)$$

$\Omega_0(\mathbf{k})$  being the unperturbed proper frequency. It is worth stressing the analogy existing between the systems described by Eq. (6c) and the *cavitating gases*,<sup>13</sup> implemented by a resonant pressure wave, on a liquid with dissolved gas. Due to the steady nature of the process, bubbles of gas are formed at the nodes of the pressure wave. The radius of each bubble follows the periodic change in time of the pressure. An accumulation of energy in the bubble is observed, under certain critical conditions, leading the bubble radius to "collapse" from sizes of about 40 m to about 0.5  $\mu\text{m}$ . Accordingly, the internal temperature is suddenly raised up to  $10^4$  K. A theoretical picture of the process can be given in terms of classical fluids' equations. The analogy between the cavitation

effect and the present theory will become even more manifest in Sec. VI, where Eq. (6c) will be applied to a continuum lattice model.

While Eq. (6c) refers to *extended* normal modes, a similar description can be applied to *localized* modes too, provided their localization length  $\lambda$  is large compared to the interatomic distance. In this case, it still makes sense to introduce a density of particles  $\rho(\mathbf{R}, t)$ , in a spatial region of linear size  $\lambda$ , around the position  $\mathbf{R}$ , where the elastic mode is localized. The application of Grüneisen's theory yields, in this case

$$\Omega(\mathbf{R}, t) \cong \Omega_0(\mathbf{R}) \left[ 1 + \gamma(\mathbf{R}) \frac{\Delta\rho(\mathbf{R}, t)}{\rho_0(\mathbf{R})} \right], \quad (7)$$

for the proper frequency of the localized mode. Note that in Eq. (7) both  $\gamma$  and  $\rho_0$  have been assumed (through the dependence on  $\mathbf{R}$ ) as *locally* defined quantities, since the material is expected to be nonhomogeneous, for *localized* modes to exist.

Equation (7) may be relevant for systems containing *mobile* atoms, such as liquids, hydrogenated metals, ionic conductors, for which the local density  $\rho(\mathbf{R}, t)$  may change in time because of *internal* atomic motions, even in the absence of external fields. Clearly, the fluctuations originated by these “internal” strains are necessarily *disordered*, while those originated by external fields, under experimental control, can be periodic by choice. Hydrogen-rich dots in hydrogenated metals, like those envisaged in Refs. 5–7, appear good candidates for the application of Eq. (7), since they are relatively large defects, corresponding to localized elastic modes, whose frequency follows the fluctuations in time of the hydrogen density in each dot.

Beside Grüneisen's theory, leading to Eqs. (6c) and (7), more complicated cases could be realized by time-dependent electromagnetic fields, if the material is strongly *piezoelectric* or *magnetostrictive*. These effects, however, are strongly model dependent, while Eqs. (6c) and (7) are “universal,” within the limits of Grüneisen's approximation.

#### IV. DISSIPATION EFFECTS: LOW DENSITY OF $s$ PHONONS

The formal analogy [Eq. (2)] underlying the preceding arguments, is obviously based on the assumption that the unperturbed potential in Eq. (1) is harmonic. It is only under this approximation that the exponential storage of energy can be described in the ideal scheme proposed above. In a real system, anharmonicity is just responsible for thermalization (or dissipation).<sup>14</sup> Thermalization makes the energy of the thermal modes (that we call “ $T$  modes”) fluctuate around the equilibrium value, and transforms the initial energy of a nonthermal phonon into thermal energy. What happens to the  $s$  modes, adsorbing energy also from the time-dependent strains, is better understood in terms of the corresponding *phonons*. In the classical limit, the amplitude  $Q(t)$  of any normal mode can be easily related to the average number  $\langle n(t) \rangle$  of phonons, since  $\langle n(t) \rangle = \langle Q^2(t) \rangle m \Omega_0 / \hbar$ , with  $m$  = particle mass. For  $T$  modes, the time average is equivalent to a thermal average, that is  $\langle n_T(t) \rangle = \kappa T [\Omega_0 \hbar]^{-1}$ . For  $s$  modes, the amplitude  $Q_s(t)$  can be set equal to  $Q_T(t) \exp(\omega_s t) / \sqrt{\alpha}$ , in the absence of dissipation, where the

pre-exponential factor  $Q_T$  behaves like a  $T$ -mode amplitude. The numerical factor  $\alpha > 1$  accounts for the fact that, depending on the initial conditions, only a fraction (that we denote as  $1/\sqrt{\alpha}$ ) of the initial amplitude does contribute to the exponential *increase*, while the remaining fraction  $1 - 1/\sqrt{\alpha}$  refers to the exponentially *decreasing* component of the motion. The importance of  $\alpha$  will become manifest in Sec. VI. If the rate of increase  $\omega_s$  is small compared to the proper frequency  $\Omega_0$  (weakly enhanced oscillations) the “fast” factor  $Q_T$  fits the thermal average, so that the *total* average number of phonons in the  $s$  mode reads  $\langle n_{\text{tot}}(t) \rangle = \kappa T [\alpha \Omega_0 \hbar]^{-1} \exp(2\omega_s t)$ , and the number of *excess* phonons (that we call  $s$  phonons), produced by the time-dependent strain, turns out to be

$$\langle n_s(t) \rangle = \frac{\kappa T}{\hbar \Omega_0} \left[ \frac{e^{2\omega_s t}}{\alpha} - 1 \right] \quad \text{for } t > t_m = \frac{\text{def } \ln \alpha}{2\omega_s} \quad (8)$$

$$= 0 \quad \text{for } t < t_m.$$

Equation (8) is actually an approximation, replacing a smooth transition to the long-time behavior, occurring around the time scale  $t_m$ , with a sharp transition occurring just at  $t_m$ . This amounts to neglect the details of the initial steps of the process, during which the decreasing and increasing components of the amplitude are comparable in magnitude. From Eq. (8), the differential equation for  $\langle n_s \rangle$  in the absence of dissipation is

$$\frac{d\langle n_s \rangle}{dt} = 2\omega_s (\langle n_s \rangle + \langle n_T \rangle) \quad (t > t_m), \quad (9a)$$

with  $\langle n_s(t=t_m) \rangle = 0$ . Note that Eq. (9a) is completely *deterministic*, since it has been obtained from the solution itself, as deduced from Eq. (1). In particular, Eq. (9a) refers to an *intensive* quantity like  $\langle n_s \rangle$  (the number of  $s$  phonons in a single  $s$  mode). In contrast, dissipation effects are *probabilistic* in nature. As discussed in Appendix A, the resulting fluctuations decrease with the square root of the number of phonons themselves, so that a deterministic equation including the dissipative effects does make sense only for the *total* number  $N_s(\omega_s, t)$  of  $s$  phonons with frequency  $\omega_s$  in the system. Setting  $N_s(\omega_s, t) = M(\omega_s) \langle n_s \rangle$ , with  $M(\omega_s)$  the number of  $s$  modes in the system (an *extensive* variable), the equation determining  $N_s(\omega_s, t)$  is obtained from the balance between the (deterministic) rate of creation  $2\omega_s$  in Eq. (9a), and the (probabilistic) rate of single-phonon decay  $\omega_1(T)$ , including all anharmonicity effects (phonon-phonon interaction, phonon-electron interaction in metals, scattering on defects, scattering on the surface,...). The resulting balance equation is

$$\frac{dN_s}{dt} = [2\omega_s - \omega_1(T)] \left( N_s + M \frac{\kappa T}{\hbar \Omega_0} \right) \quad (t > t_m). \quad (9b)$$

Until the number of  $s$  phonons per unit volume is small, one can neglect the effects of their scattering on the actual temperature (the opposite case is considered in Sec. V), so that the temperature in  $\omega_1(T)$  is constant, and fixed only by the external conditions. The solution of Eq. (9b), in the same approximation leading to Eq. (8), is immediately found as

$$N_s = M \frac{\kappa T}{\hbar \Omega_0} \left[ \frac{e^{(2\omega_s - \omega_1)t}}{\alpha} - 1 \right] \quad \text{for } t > t_{\text{diss}} \stackrel{\text{def}}{=} \frac{\ln \alpha}{2\omega_s - \omega_1}, \quad (9c)$$

$$= 0 \quad \text{for } t < t_{\text{diss}}.$$

The initial ‘‘waiting time’’  $t_{\text{diss}}$ , including the dissipative effects, now replaces  $t_m$  appearing in Eq. (8). The condition  $N_s > 0$  [first Eq. (9c)] yields

$$\omega_{\text{eff}}(T) = 2\omega_s - \omega_1(T) > \frac{\ln \alpha}{t}. \quad (10)$$

If condition (10) is satisfied,  $\omega_{\text{eff}}(T)$  is the effective rate of exponential increase of the  $s$  phonons’ number. If, instead,  $\omega_{\text{eff}}(T)$  is negative or zero,  $N_s$  remains equal to zero at any time.

Once condition (10) is satisfied, the steady time fluctuation of the proper frequency does produce an exponential divergence of the  $s$  mode’s amplitude, even in the presence of *single-phonon* absorption. Of course, other mechanisms are expected to prevent the divergence of the oscillation amplitude. In particular, the next step of the perturbative approach is to account for all channels involving the *coherent* scattering of  $s$  phonons. These channels may include a number of high-energy processes (local structural changes, vacancy formation, activation of optical modes), which would be normally forbidden to  $T$  phonons. Introducing a rate  $\omega_j(T)$  ( $j \geq 1$ ) for the coherent adsorption of  $j$  phonons, the total rate of dissipation for  $s$  phonons becomes

$$\omega_{\text{tot}}(\langle n_s \rangle) = \sum_{j=1}^{\langle n_s \rangle} j \omega_j, \quad (11)$$

replacing  $\omega_1$  in Eq. (9b). However, Eq. (9b) makes sense only if the total number of scattered phonons can be taken as a *differential* quantity in the lhs. As shown in Appendix A, this can be done only in a limiting case, that does not differ too much from the discussion reported above for the single-phonon process  $\omega_1$ . In contrast, a remarkable difference exists when there is a *resonant* channel for the coherent adsorption of  $n^*$  ( $\geq 1$ ) phonons, leading to a *discontinuous* behavior of  $\omega_{\text{tot}}$ . For instance, let

$$\omega_{\text{tot}} = \omega_1 \quad \text{for } \langle n_{\text{tot}} \rangle < n^*$$

$$\omega_{\text{tot}} = \omega_1 + n^* \omega_{n^*} \quad \text{for } \langle n_{\text{tot}} \rangle \geq n^*, \quad (12a)$$

where  $\langle n_{\text{tot}} \rangle = \langle n_s \rangle + \langle n_T \rangle$  is the *total* number of phonons in the  $s$  mode ( $s$  phonons +  $T$  phonons). The problem comes from noticing that the resonant absorption channel is inactive, for any *single* mode, until the total number of phonons  $\langle n_{\text{tot}} \rangle$  is less than  $n^*$ . If  $2\omega_s$  is smaller than the minimum rate  $\omega_1$ , the average number of  $s$  phonons in the whole system is obviously zero. If  $2\omega_s$  is larger than the maximum rate  $\omega_1 + n^* \omega_{n^*}$ , the average number of  $s$  phonons in *each*  $s$  mode increases exponentially in time with decreasing fluctuations, as shown in Appendix A. Instead, the condition

$$\omega_1 < 2\omega_s < \omega_1 + n^* \omega_{n^*} \quad (12b)$$

makes it necessary to split in time the two cases  $\langle n_{\text{tot}} \rangle < n^*$  and  $\langle n_{\text{tot}} \rangle \geq n^*$  for any *single* mode. Hence, the problem of the fluctuations cannot be avoided by taking the thermody-

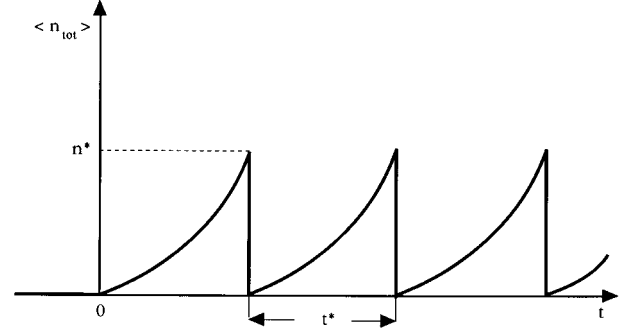


FIG. 2. Qualitative sketch of the behavior in time of the number of phonons in a *single*  $s$  mode, in the presence of a resonant adsorption process involving  $n^*$  phonons [Eqs. (12)], in the ideal limit  $n^* \omega_{n^*} \rightarrow \infty$ .

namic limit, as we did in Eqs. (9), because  $\langle n_{\text{tot}} \rangle$  is an intensive variable. The problem raised by Eqs. (12) is to calculate, at a given instant  $t > 0$ , the probability for a single  $s$  mode to contain a given number of  $s$  phonons, on account of the fact that there is a *deterministic* process producing phonons with a rate  $2\omega_s$ , and two *probabilistic* processes of dissipation, one of which is activated only when the number of phonons itself is larger than  $n^*$ . The resulting equation is rather complicated, and its general discussion is left to further investigations. The only limit case leading to a simple solution is  $n^* \gg \langle n_T \rangle \gg 1$  and  $n^* \omega_{n^*} \rightarrow \infty$ . The first condition ensures that we can approximately use Eq. (9b) for  $\langle n_s \rangle$  too, whenever  $\langle n_{\text{tot}} \rangle < n^*$  (Appendix A). This leads one to define a time interval

$$t^* = \omega_{\text{eff}}^{-1} \ln(n^* + \langle n_T \rangle) + t_{\text{diss}},$$

during which  $\langle n_s \rangle$  increases from 0 to  $n^* - \langle n_T \rangle$ . The limit  $n^* \omega_{n^*} \rightarrow \infty$  allows one to claim that the resonant adsorption of  $n^*$  phonons occurs with probability 1 in any arbitrarily small interval of time, after the activation of the resonant channel. This resets the mode to the initial state of zero phonons, and makes the ‘‘loading’’ of phonons start again, with  $t$  replaced by  $t - t^*$ . The resulting picture is thus a periodical (with period  $t^*$ ) increase of  $\langle n_{\text{tot}} \rangle$ , up to the value  $n^*$ , followed by a sharp decrease to zero, due to the resonant adsorption (Fig. 2). It is worth stressing that a physical realization of Eqs. (12) could be, for example, the periodical ‘‘pumping’’ of  $n^*$  *acoustic* phonons into the *optical* phonon branch.

## V. DISSIPATION EFFECTS FOR MANY $s$ PHONONS: SELF-CONSISTENT THERMALIZATION

The atomic displacement  $\mathbf{q}(\mathbf{r}_n, t)$  in any site  $\mathbf{r}_n$  of a crystalline lattice can be written, in the presence of  $s$  modes

$$\mathbf{q}(\mathbf{r}_n, t) = \frac{1}{\sqrt{N}} \left[ \sum_{\mathbf{k} \neq \mathbf{k}^*} \mathbf{e}(\mathbf{k}) Q_T(\mathbf{k}, t) e^{i\mathbf{k} \cdot \mathbf{r}_n} + \sum_{\mathbf{k}^*} \mathbf{e}(\mathbf{k}^*) Q_S(\mathbf{k}^*, t) e^{i\mathbf{k}^* \cdot \mathbf{r}_n} \right], \quad (13a)$$

where  $\mathbf{k}^*$  are the wave vectors of the  $s$  modes.  $Q_s(\mathbf{k}^*, t)$  and  $Q_T(\mathbf{k}, t)$  are the  $s$  modes' and  $T$  modes' amplitudes, respectively, and  $\mathbf{e}(\mathbf{k})$  is the polarization unit vector of the  $\mathbf{k}$  mode (for brevity, we do not distinguish here between transversal and longitudinal modes). Taking the square of Eq. (13a), and using the random-phase approximation (valid at high temperatures), the average on time of  $\mathbf{q}^2(\mathbf{r}_n, t)$  results from summing up the average square moduli, whence, from the discussion in Sec. IV

$$\langle \mathbf{q}^2(\mathbf{r}_n, t) \rangle \cong \mathbf{q}_T^2(T) + \mathbf{q}_s^2(T, t), \quad (13b)$$

where

$$\mathbf{q}_T^2(T) = \frac{D\kappa T}{mN} \sum_{\mathbf{k} \neq \mathbf{k}^*} \Omega_0^{-2}(\mathbf{k}) \quad (14a)$$

is the thermal part of the square amplitude ( $D$  = dimension of the system), depending on the temperature only, and

$$\mathbf{q}_s^2(T, t) = \frac{D\hbar}{mN} \sum_{\mathbf{k}^*} \frac{\langle n_s(\mathbf{k}^*, t) \rangle}{\Omega_0(\mathbf{k}^*)} \quad (14b)$$

is the ‘‘special’’ part, following the behavior in time of the  $s$  phonons. The important quantity in Eq. (14b) is actually the number of  $s$  phonons per unit volume. So far, any possible effect of the ‘‘dissipated’’  $s$  phonons on the thermal bath itself, has been neglected at all. This is possible only if the concentration of  $s$  phonons produced by the strains does remain small. Instead, suppose that the sum in Eq. (11) converges to an upper limiting value  $\omega_M(T)$  for  $\langle n_s \rangle \rightarrow \infty$ . In addition, let the number of  $s$  modes such that  $2\omega_s(\mathbf{k}^*) > \omega_M(T_i)$ , at the initial temperature  $T_i$ , be an *extensive* quantity. If so, the terms increasing exponentially at long times [with rates  $\omega_{\text{eff}} = 2\omega_s(\mathbf{k}^*) - \omega_M(T_i)$ ], yield a *finite* contribute to the sum Eq. (14b), even in the thermodynamic limit, and make the oscillation amplitudes of *all* atoms diverge in turn. In this case, the basic assumption that the  $s$  phonons do not influence the thermal bath, does not make sense any more, at long times. In order to account for the effects of the dissipated  $s$  phonons on the thermal bath, one may define the *actual* temperature  $T(t)$  at any instant, just from the square oscillation amplitude in the coordinate space, that is

$$T(t) = \frac{mN}{D\kappa} \langle \mathbf{q}^2(\mathbf{r}_n, t) \rangle \left[ \sum_{\mathbf{k}} \Omega_0^{-2}(\mathbf{k}) \right]^{-1}. \quad (15)$$

From Eqs. (13), (14), and (15), it follows that  $T(t)$  is an *increasing* function of time, if  $s$  phonons are produced. Being an increasing function of  $T(t)$  itself, the maximum rate of dissipation  $\omega_M(T(t))$  behaves accordingly [see, for example, Eq. (19) in Sec. VI]. Then the condition  $2\omega_s(\mathbf{k}^*) > \omega_M(T(t))$  for a  $s$  mode to produce  $s$  phonons, becomes more and more stringent, with increasing time (that is, with increasing number of  $s$  phonons). In practice, the larger the concentration of  $s$  phonons, the smaller the number of  $s$  modes that can produce them. Accounting for this nonlinear countereffect is what we call ‘‘self-consistent thermalization’’ of the  $s$  phonons. As we will see in the next section, the self-consistent thermalization may lead the number of  $s$

phonons [and the value of the actual temperature  $T(t)$ , Eq. (15)] to saturate at long times to finite limiting values.

## VI. THE CONTINUUM-LATTICE MODEL

In the continuum-lattice model (CLM), the frequency  $\Omega = c(\rho)k(\rho)$  of each normal modes involves the sound velocity  $c(\rho)$  and the wave vector's modulus  $k(\rho)$ , both depending on the homogeneous density  $\rho$ . In particular, in three dimensions  $k(\rho) = k(\rho_0) \sqrt[3]{\rho_0/\rho}$ ,  $\rho_0$  being a reference value, that we take as the unperturbed density. The expression for  $k(\rho)$  simply follows from the boundary conditions, according to the discussion in Sec. III. At this stage, we can set  $k(\rho_0) = k$ , with the implicit assumption that  $k$  refers to the *unperturbed* system, so that its upper limiting value  $k_D$  (the Debye wave vector) is *independent* of time. Equation (6c), describing the effect of a uniform time-dependent pressure in Grüneisen's approximation, can be now applied to the CLM, with the result<sup>15</sup>

$$\Omega(k, t) = c_0 k \left[ 1 + \gamma \frac{\Delta\rho(t)}{\rho_0} \right], \quad (16a)$$

for  $k < k_D$ . The quantity  $c_0$  is the unperturbed sound velocity (for simplicity, we use the same formalism both for *transversal* and for *longitudinal* modes). From Eq. (16a) one can define a time-dependent sound velocity  $c(t) = c_0 + \Delta c(t)$ , such that

$$\Delta c(t) = c_0 \gamma \frac{\Delta\rho(t)}{\rho_0} \leq \Delta c_M. \quad (16b)$$

With  $\Delta c(t)$  behaving like the model fluctuation in Fig. 1, Eqs. (5) and (16b) readily yield

$$k^* = k_j = j\pi(\tau c_0 + \Delta\tau \Delta c_M)^{-1} \quad (j=0,1,2,\dots), \quad (17a)$$

$$\omega_s(k_j) \cong \frac{\xi}{\tau} |\sin(j\pi \Delta\tau/\tau)|, \quad (17b)$$

where

$$\xi \stackrel{\text{def}}{=} \frac{\Delta c_M}{c_0} \quad (17c)$$

is the smallness parameter of the problem. In particular, the special choice  $\Delta\tau/\tau = 1/2$  (which makes Fig. 1 as similar as possible to a sin-cos perturbation) leads to a further simplification, since Eqs. (17) become

$$k_j \cong \frac{(2j+1)\pi}{\tau c_0} (1 - \xi/2); \quad (j=0,1,2,\dots), \quad (18a)$$

$$\omega_s(k_j) \cong \xi/\tau = \omega_s \quad (\text{independent of } j), \quad (18b)$$

so that  $\omega_s$  is the *same* for all the  $s$  modes, whose wave vectors satisfy the quasiresonance condition (18a). From now on, we will neglect the small term  $\xi$  in Eq. (18a). If we are interested in condition (10), marking the onset of any macroscopic effect related to the existence of nonthermalized  $s$  phonons, we need an explicit formula for the rate of decay  $\omega_1$  of a *single* phonon of wave vector  $\mathbf{k}$ , into  $p(>1)$  different phonons. For a *nonmetallic* solid at high temperatures

( $T >$  Debye temperature),  $\omega_1$  is usually calculated as the sum of three terms, that is, normal and umklapp processes with  $p=2$ , and normal processes with  $p=3$ .<sup>16</sup> The resulting expression reads as follows:

$$\omega_1(k, T) = 2\gamma^2\omega_D \left\{ \left[ \frac{T}{T_0} + A \left( \frac{T}{T_0} \right)^2 \right] \left( \frac{k}{k_D} \right)^2 + 30\pi^2 \frac{T}{T_0} \left( \frac{k}{k_D} \right)^4 \right\}, \quad (19)$$

where  $\gamma$  is the Grüneisen parameter,  $\omega_D = c_0 k_D$  is the Debye frequency,  $T_0 = mc_0^2/\kappa$  is a characteristic temperature, and  $A$  is a geometric factor somewhat larger than 1. On setting  $k = k_j$  in Eq. (19), with  $k_j$  given by Eq. (18a), condition (10) yields a lower limiting value for  $\tau$ , below which *all*  $s$  phonons are thermalized, even for the lowest possible value  $k_0$  of the quairesonant wave vector. In practice, condition (10) *necessarily* implies that

$$\frac{\tau\omega_D}{2\pi} > \frac{\gamma^2\pi}{2\xi} \left[ \frac{T}{T_0} + A \left( \frac{T}{T_0} \right)^2 \right], \quad (20a)$$

provided

$$\frac{\tau\omega_D}{2\pi} \gg 1. \quad (20b)$$

In general, if  $T/T_0 < 10^{-1}$  (that means, typically,  $T < 10^3$  K), even the second term in square brackets in Eq. (20a) can be neglected, as we will do from now on. Condition (10) also yields an upper limiting value for the wave vectors satisfying Eq. (18a):

$$k_j < k_D \left[ \frac{T_0}{\gamma^2 T \omega_D} \left( \frac{\xi}{\tau} - \frac{\ln \alpha}{2t} \right) \right]^{1/2} \stackrel{\text{def}}{=} k_M(T, t). \quad (20c)$$

Since, from conditions (20a) and (20b),  $\tau$  is large (but not arbitrarily large: see below), it is reasonable to approximate the discrete set of values determined by Eq. (20c) with a *continuous band* of  $k$  values, ranging from  $k_0$  to  $k_M$  (see Appendix B). If now we neglect any coherent scattering effect, on setting  $\omega_1 = \omega_M$ , the self-consistent thermalization can be approached as follows: Eq. (9c) yields the number of  $s$  phonons (proportional to the *initial* temperature  $T_i$ ), for any  $k_j$  ranging from  $k_0$  to  $k_M$ . On inserting Eq. (9c) into Eq. (14b), and on transforming the sum into an integral, the  $s$  part of the square oscillation amplitude is obtained as a functional of  $\omega_1(T, k^*)$ , where  $T$  is now taken as the (unknown) *actual* temperature. By means of Eq. (14a) (with  $T$  replaced by  $T_i$ ), the total square amplitude is obtained, as a function of  $T$ , from Eq. (13b). The resulting expression can be put into Eq. (15), that becomes the evolution equation for the actual temperature itself (Appendix B):

$$T(x) = T_i \left\{ 1 + \frac{\xi k_0}{\pi k_D F(T)} \left[ \frac{e^{-x^2}}{f(x)} \int_{f(x)F(T)}^x e^{-y^2} dy - 1 + F(T) \right] \right\}, \quad (21a)$$

where

$$x = (2\omega_s t - \ln \alpha)^{1/2}; \quad f(x) = x \left( 1 + \frac{\ln \alpha}{x^2} \right),$$

$$F(T) = k_0/k_M(T, \infty) = \text{from Eq. (20c)} = \gamma \frac{k_0}{k_D} \left( \frac{T\omega_D\tau}{T_0\xi} \right)^{1/2}. \quad (21b)$$

At *constant volume*,<sup>17</sup>  $k_M(T, \infty)$  is the only quantity depending explicitly on  $T(t)$ , while at constant pressure both  $k_D$  and  $k_0$  would be explicit functions of  $T(t)$ , through the specific volume and  $c_0$ , respectively. Limiting ourselves to processes at constant volume, the solution of Eq. (21a) can be given analytically in the limit  $x \rightarrow \infty$  ( $t \rightarrow \infty$ ) as

$$T(t) \cong T_\infty \left[ 1 - \frac{\ln(2\omega_s t)}{2\omega_s t} \right], \quad (22)$$

with

$$T_\infty = \xi T_0 \frac{\omega_D\tau}{(\gamma\pi)^2} \quad \text{for } \tau < L/c_0. \quad (23a)$$

The condition  $\tau < L/c_0$  in Eq. (23a) is equivalent to assume that  $k_0$  [Eq. (18a)] is larger than the minimum  $k$  value  $k_m \cong \pi/L$  for a system of linear dimension  $L$ . Otherwise, the lower limit  $f(x)k_0/k_M$  of the integral in Eq. (21) must be replaced by  $f(x)k_m/k_M$ . The preceding calculations for the self-consistent thermalization do not change, but the value of the saturation temperature  $T_\infty$  becomes *size dependent*:

$$T_\infty = \xi T_0 \frac{k_D L^2}{(\gamma\pi)^2 c_0 \tau} \quad \text{for } \tau > L/c_0. \quad (23b)$$

The saturation value  $T_\infty$  obviously coincides with the temperature at which the inequality (20a) becomes an identity (apart from the second term in square brackets, which has been neglected). Physically, Eqs. (23) indicate that the system, initially at the temperature  $T_i$ , is driven by the periodic stress to a higher temperature  $T_\infty$  (independent of  $T_i$ ), with a smooth asymptotic evolution  $\ln(t)/t$  (see Fig. 3). The period  $\tau$  of the perturbation cannot be arbitrarily large, if  $s$  phonons have to be produced in the initial steps of the process. In fact, the *finiteness* of the system yields a size-dependent upper limiting value  $\tau_M$  for  $\tau$ . This follows from requiring that, at the initial time, the maximum value  $k_M(T_i)$  of the wave vectors corresponding to  $s$  phonons, is larger than the minimum value  $k_m \cong \pi/L$ . From Eq. (20c) (with  $T = T_i$ ), this condition reads

$$\tau < \tau_M \stackrel{\text{def}}{=} \frac{\xi T_0 \omega_D}{\gamma^2 T_i} \left( \frac{L}{c_0 \pi} \right)^2. \quad (24a)$$

A lower limiting value  $\tau_m$  for  $\tau$  follows from Eq. (23a), on requiring that  $T_i < T_\infty$ :

$$\tau > \tau_m \stackrel{\text{def}}{=} \omega_D^{-1} (\gamma\pi)^2 \frac{T_i}{T_0 \xi}. \quad (24b)$$

If one of the conditions (24) is violated, no  $s$  phonons are present in the system, and the actual temperature coincides with the initial temperature. From Eqs. (23), it is easy to see that

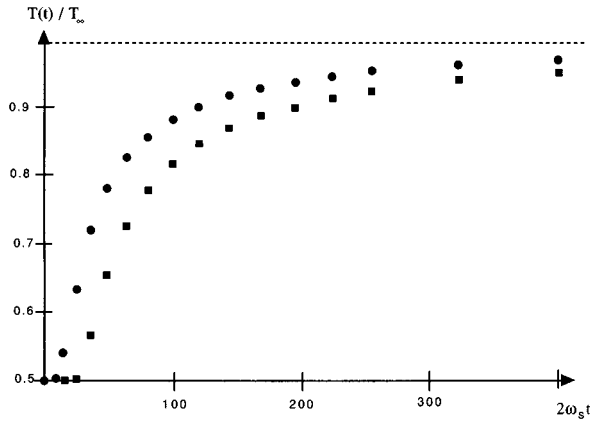


FIG. 3. Numerical solution of Eqs. (21) [see also Eq. (B5)] for the actual temperature  $T(t)$  in a continuum lattice, under the action of a time-dependent uniform stress as in Fig. 1. The saturation temperature  $T_\infty$  is taken twice as large as the initial temperature  $T_i$ . Two plots are reported, for  $\Gamma=10^{-3}$  (circles) and for  $\Gamma=10^{-6}$  (squares). Note the steep increase at times of order  $10/\omega_s$ , and the slow saturation at  $T_\infty$ . The numerical data are practically insensitive to the specific value of  $\alpha$ , for  $\alpha$  of order unity.

$$T_\infty \leq T_M \stackrel{\text{def}}{=} \xi T_0 \frac{\omega_D L}{c_0 (\pi \gamma)^2}. \quad (25)$$

The upper limiting value  $T_M$  of the saturation temperature corresponds to  $\tau=L/c_0$ , at which the two expressions (23a) and (23b) do coincide. The full dependence of  $T_\infty$  on  $\tau$  is sketched in Fig. 4, under the obvious condition  $T_i < T_M$ , which yields, from Eq. (25)

$$\xi > (\pi \gamma)^2 \frac{T_i c_0}{T_0 \omega_D L}. \quad (26)$$

The opposite case  $T_M < T_i$  is not excluded mathematically, but it simply means that the dissipation rate is always larger than the production rate of  $s$  phonons, so that the temperature remains at the initial value  $T_i$ , and the average number of  $s$  phonons is zero. Recalling Eqs. (17c) and (16b), condition (26) becomes

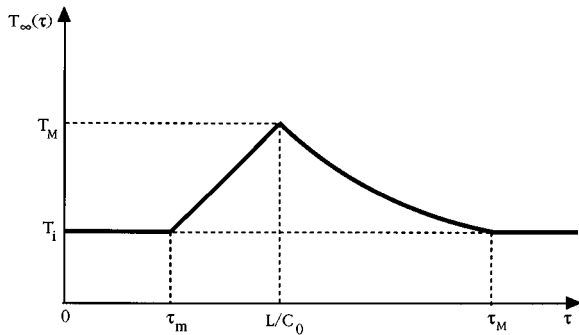


FIG. 4. Dependence on  $\tau$  (the period of the stress) of the saturation temperature  $T_\infty$  for a continuum lattice model of linear size  $L$  [Eqs. (29b)]. Note the “window”  $[\tau_m, \tau_M]$  of allowed values of  $\tau$ , giving rise to an increase of the actual temperature above the initial value [see Eqs. (24)].

$$\frac{\Delta \rho_M}{\rho_0} > \pi \gamma \frac{T_i a}{T_0 L}, \quad (27)$$

where  $a$  is the interatomic distance, so that  $\omega_D = c_0 k_D \equiv c_0 (\pi/a)$ . Inequality (27) provides the (size-dependent) lower limit on the maximum density fluctuation, in order that there are macroscopically detectable  $s$  phonons. In the classical limit, the state equation of CLM reads

$$P(T, \rho) = 3 \gamma \rho \left( \kappa T - \frac{3}{8} \hbar \omega_D \right),$$

so that the lower limit for the maximum pressure change  $\Delta P_M$  is readily obtained from Eq. (27):

$$\frac{\Delta P_M}{P(T_\infty, \rho_0)} > \pi \gamma \frac{T_i a}{T_0 L}. \quad (28)$$

Since the pressure of solids at relatively high temperatures is of order  $10^5$  atm, Eq. (28) tells us that for a *pure, nonmetallic monocystal*, of cubic form, with sides about 1 cm long, at initial temperature of some hundred Kelvin, the periodic pressure change must exceed a value of about  $10^{-2}$  atm ( $\gamma \approx 2$ ), in order that  $s$  phonons can raise the actual temperature up to macroscopically detectable values. This threshold value for the pressure might look surprisingly low, since  $10^{-2}$  atm corresponds to nothing but a delicate finger touch. However, one should now recall that the theory, in the present form, holds only for *adiabatic* strains. As stressed in Sec. III, this means that the frequency of the strain fluctuation must be small compared to the *minimum* proper frequency of the normal modes. For an ideal solid of linear size  $L$ , this condition implies  $\tau \gg L/c_0$ , which shifts the region of applicability of the theory well across the top of the peak in Fig. 4. The expression to be accounted for is now Eq. (23b), showing that the condition for  $T_\infty$  to be larger than  $T_i$  actually reads

$$\frac{\Delta P_M}{P(T_\infty, \rho_0)} \gg \pi \gamma \frac{T_i a}{T_0 L} \quad \text{for } \tau \gg L/c_0. \quad (29)$$

Instead, Eq. (28) refers to the case  $\tau \approx L/c_0$ , that makes the strain nonadiabatic for the low-frequency modes of the system. As stressed in Sec. III, the adiabaticity condition  $\tau \gg L/c_0$  is equivalent to assuming that the time-dependent perturbation does produce anharmonic effects only. In this case one is sure that the predicted temperature increase is entirely due to the  $s$  phonons, and not to the thermalization of  $T$  phonons produced by viscoelastic effects.

Now we wish to stress an apparent paradox emerging from Eqs. (23b) and (24): since  $\xi$  is *linear* in the Grüneisen parameter  $\gamma$  [Eq. (17c)] the limit  $\gamma \rightarrow 0$  yields  $T_\infty \rightarrow \infty$ ,  $\tau_M \rightarrow \infty$  and  $\tau_m \rightarrow 0$ . Thus a perfectly elastic lattice would be able to store an arbitrarily large amount of  $s$  phonons, for any value of the perturbation's period. But a perfectly elastic lattice cannot contain  $s$  phonons by definition (Sec. III). The problem looks unescapable, for it has a first-principles origin: the exponential increase of the number of  $s$  phonons is a *first-order* effect in  $\gamma$ , while the contrasting dissipation rate [Eq. (19)] is a *second-order* effect. The key for the solution is just the parameter  $\alpha$ . As reported in the caption of Fig. 3, the value of  $\alpha$ , unless not too large, is irrelevant for the data



in Fig. 3, obtained from *finite* (though small) values of  $\xi$ . But the limit  $\gamma \rightarrow 0$  also yields  $t_{\text{diss}} \rightarrow \infty$  [Eq. (9c)], even for  $\alpha$  arbitrarily close to 1. Equations (23b) and (24) have been deduced under the implicit condition  $t/t_{\text{diss}} \rightarrow \infty$ , that is, over time scales large compared to the initial waiting time interval  $t_{\text{diss}}$ , during which the behavior of the harmonic oscillator's amplitude is not still exponential. With  $t_{\text{diss}}$  diverging, the system does not contain  $s$  phonons at all, and the limit of the perfectly elastic CLM is recovered. Note that the condition  $\alpha = 1$ , for which  $t_{\text{diss}} = 0$  anyway, is statistically irrelevant, since it corresponds to a zero-measure set of (classical) initial conditions. In a forthcoming paper, concerned with the *quantum* oscillator with time-dependent frequency, we will show that  $\alpha = 2$  is the suitable value for the problem under consideration. However, the ‘‘paradox’’ discussed above points out an interesting result: the waiting time for the  $s$  phonons' production does actually increase with increasing elasticity, but the *intensity* of the resulting effects does increase in turn, when the coherent production finally starts. This could have relevant consequences in view of more realistic approaches to the anharmonicity, introducing a temperature dependence in the Grüneisen parameter.

Of course, the preceding formulas for CLM are far from reliable, when applied to real cases. Besides the phonon-phonon scattering accounted for in the ideal CLM, any kind of defects (grain boundaries, dislocations, atomic impurities, surface scattering, free electrons in metals) would contribute their own terms to the dissipation rate. Hence, for  $s$  phonons to produce measurable effects, one should expect to find more stringent conditions than those predicted in the present ideal case. However, the low value of the ‘‘ideal’’ threshold pressure [Eq. (28)] provides an encouraging indication, in view of a real experimental test.

## VII. SUMMARY AND CONCLUSIONS

The problem discussed in the preceding sections can be summarized as the application of the swing-with-child Eq. (1) to elementary elastic excitation in solids, in the presence of dissipation effects. The general solution of Eq. (1) has been given in Sec. II, by mapping Eq. (1) into a one-dimensional Schrödinger equation [Eq. (2)]. The cases a of disordered and b periodic fluctuations of the proper frequency have been considered separately. However, the presence of dissipation makes the two cases coincide in b, for many practical purposes, with the disordered fluctuation replaced by a ‘‘coherent’’ periodic function. The crucial point is the existence of special frequencies, at which the elastic modes' amplitude diverge exponentially in time (in the absence of dissipation). These special modes are indicated as ‘‘ $s$  modes,’’ while their *excess* phonons (that is, the phonons generated by the time-dependent strain), are denoted as  $s$  phonons. Some physical applications of Eq. (1) have been discussed in Sec. III, with special reference to what we call an *adiabatic* time-dependent strain, acting on the proper frequencies of normal modes in condensed systems. Adiabaticity means in general that the frequency spectrum of the strain fluctuation (obtained by Fourier time transforming) lies well *below* the normal modes' spectrum of the system, with sufficiently small amplitudes. In this case, there is no way for the strain energy to enter the phonon bath, except Eq. (1),

since *viscoelastic* effects (coupling the normal modes to the strain velocity) are made negligible. Equation (1) is thereby assumed as a pure *anharmonic* effect, to which Grüneisen's theory provides the most elementary and ‘‘universal’’ approach. Explicit formulas are obtained for *extended* modes in crystals, whose proper frequencies can be made time dependent by *periodic external* stresses. Similar expressions for *localized* modes in nonhomogeneous materials do apply as well, but the proper frequencies' changes are now attributed to *disordered* fluctuations of the local density, due to *internal* ionic motions.

In Sec. IV, the dissipation effects are considered in the case of low  $s$  modes' concentration. This means that the ionic oscillations are only negligibly influenced by the  $s$  phonons. Once stressed that the creation of  $s$  phonons is a *deterministic* process, while their absorption is *probabilistic* in nature, a detailed-balance equation can be written for the *total* number of  $s$  phonons present in the sample, provided the number of  $s$  modes which they originate from is an extensive variable. This can be easily done for single-phonon scattering processes, whereas the inclusion of multiphonon processes may considerably complicate the matter. In particular, in the case of Eqs. (12), a resonant absorption channel, activated only when the number of phonons in a single mode is larger than  $n^*(\geq 1)$ , cannot be turned into a detailed-balance equation, but requires a statistical approach based on the single-mode behavior. In the ideal case of a diverging resonant rate of absorption, it is possible to guess that the population of phonons in each  $s$  mode oscillates periodically between 0 and  $n^*$ , as sketched in Fig. 2.

In Sec. V, we account for the influence of the  $s$  phonons on the ionic oscillations. In this case the evolution of the system is described by the so-called ‘‘self-consistent thermalization’’ of the  $s$  phonons: if  $s$  phonons are dissipated in the system, the *actual* temperature  $T(t)$  and the maximum rate of dissipation  $\omega_M(T(t))$  are both *increasing* functions of time. This leads the concentration of  $s$  modes producing  $s$  phonons to *decrease* in time, so that a nonlinear counter-effect is produced, making the actual temperature saturate at a finite limiting value  $T_\infty$  (Fig. 3). It should be noticed that the *actual* temperature  $T(t)$  is quite different from the *thermodynamic* temperature one would obtain simply by heating the system. In the latter case each oscillator's energy would be characterized by a Poisson distribution, whose variance increases linearly with the temperature. This is an obvious consequence of the chaotic approach to the energy equipartition, where each microscopic oscillator is a canonic system exchanging heat with its neighbors. The actual temperature  $T(t)$ , instead, is merely a thermodynamic measure of a *non-chaotic* increase of the number of  $s$  phonons, due to an external perturbation. The only chaotic element is the initial temperature  $T_i$  [Eq. (21)]. Indeed, the saturation temperature  $T_\infty$  is independent of  $T_i$  [Eqs. (22)], so that the energy of each oscillator tends to the asymptotic value, not only in average, but with a distribution converging to  $\delta(E - \kappa T_\infty)$ . This is a fundamental consequence of the coherent (nonergodic) production of  $s$  phonons from the time-dependent strain. The oscillators in real space can be now regarded to as microscopic systems whose energy increment becomes more and more *coherent*, with increasing time and with increasing actual temperature. This opens, in principle, the possibility of

a time-controlled, coherent-phase transition, if the system can exist in two allotropic phases  $A$  and  $B$ . The resulting effects are expected to be very similar to those obtained in a cavitating gas.<sup>13</sup> In fact, suppose we apply the CLM calculations at constant volume (Sec. VI) to the system in the phase  $A$ . It is conceivable that in some conditions, a critical value  $T_c$  of the actual temperature does exist, between  $T_i$  and  $T_\infty$ , at which the increasing internal pressure makes the system undergo a structural phase transition from  $A$  to  $B$ . Since  $T_c < T_\infty$ , the critical temperature is reached in a *finite* time. For  $T_c$  sufficiently close to  $T_\infty$ , the critical pressure is attained by all parts of the system *simultaneously*. In the resulting *coherent*-phase transition, part of the *macroscopic* elastic energy loaded into the system for reaching  $T_c$  would be coherently released, (for example, by a sudden contraction of the proper volume), and a shock-wave like those envisaged in Refs. 5–7, and like those observed in the critical cavitation, might be produced. If the self-consistent thermalization of  $s$  phonons could be repeated for phase  $B$  too, in such a way that the system can be driven again to the original phase  $A$ , a steady, periodical emission of shock waves would be produced. The results obtained for the *ideal* CLM, with only anharmonic channels of scattering, indicate that the threshold pressure fluctuation, for the self-consistent thermalization to produce macroscopic effects, is size dependent and may be very low, for macroscopic monocrystals. This looks encouraging, in view of the experimental verification of the predicted effects.

Another aspect which we can only allude to, is the electron-phonon interaction in the presence of  $s$  phonons, a field that has not been considered at any level, in the present paper, but actually deserves some attention.

*Note added in proof.* The analogy between Eq. (1) and the Schrödinger equation was stressed by L. P. Pitaevski [see L. Landau and E. Lifshitz, *Mécauque* (MIR Editions, Moscow, 1969), footnote on pp. 214–215.

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#### APPENDIX A

We first recall here the conditions of applicability for a time-dependent equation based on the concept of *probabilistic* transition rate  $\omega$  (for any process). With a fixed arbitrary time interval  $\Delta t \ll \omega^{-1}$ , and a number  $N$  of incoherently processing systems, the probability  $P(\Delta N)$  that  $\Delta N$  of them do actually perform the process in the time interval  $\Delta t$  is given by the binomial distribution

$$P(\Delta N) = \frac{N!}{\Delta N!(N-\Delta N)!} p^{\Delta N} (1-p)^{N-\Delta N}, \quad (\text{A1})$$

with  $p = \omega \Delta t$ . Note that the process can be a decay (absorption), or a *duplication* of the system itself (creation). By

maximizing  $\ln P(\Delta N)$  with respect to  $\Delta N$ , the binomial distribution can be approximated by a Gaussian, close to the extremant  $\Delta \bar{N} = N \omega \Delta t$ . Treating  $x = \Delta N/N$  as a continuous variable, the resulting probability distribution reads

$$P(x) \cong \sqrt{\frac{N}{p}} \exp\left[-\frac{N(x-p)^2}{p}\right] \left[ \int_{-\sqrt{Np}}^{\infty} e^{-z^2} dz \right]^{-1}, \quad (\text{A2})$$

that, in the limit  $N \rightarrow \infty$ , tends to  $\delta(x-p)$ . Thus, in the same limit, one gets  $x = p = \omega \Delta t$  as the *unique* possibility of realization. On taking  $\Delta t \rightarrow 0$ , this yields the usual *deterministic* equation  $\dot{N} = \pm \omega N$  (with  $+$  and  $-$  corresponding to creation and absorption, respectively). It is thereby clear that the deterministic nature of any equation based on the concept of probabilistic transition rate depends on the large- $N$  limit. The same limit is also consistent with the further assumption of treating  $\Delta N$  (a non-negative integer), as a differential quantity. For *adsorption* processes, the large- $N$  limit necessarily corresponds to the thermodynamic limit. It is only for an extensive variable that an expression like  $N(t) = N(0) \exp(-\omega t)$  can make sense even at long times, since  $N(0)$  can be taken as an arbitrarily large quantity. In contrast, for *creation* processes, an expression like  $N(t) = N(0) \exp(\omega t)$  becomes more and more correct, just with increasing time, even if  $N(0)$  is *not* arbitrarily large. In this case, in fact, the exponential increase itself makes the fluctuations become negligible at long times. For example, if condition (10) is satisfied, Eq. (9b) can be indifferently referred to the  $s$  phonons in a *single*  $s$  mode, or to *all* the  $s$  phonons (of the same kind) present in the system. In the opposite case, Eq. (9b) has to be intended as determining the average behavior of all  $s$  modes in the system, while the behavior of the single  $s$  mode could display relevant fluctuations.

The preceding discussion is useful when  $\omega_1$  is replaced by  $\omega_{\text{tot}}$  [Eq. (11)], and the coherent absorption of  $j$  phonons is formally accounted for. As soon as we wish to include processes in which the expected variation may equal the number of existing phonons, the fluctuations between the single  $s$  mode's behavior and the average behavior described by Eq. (9b) might become relevant from the physical view point. No problem exists if

$$2\omega_s > \sum_{j=1}^{\infty} j \omega_j \stackrel{\text{def}}{=} \omega_M. \quad (\text{A3})$$

In this case the rate of creation of  $s$  phonons is larger than any rate of scattering, and the long-time behavior predicted by Eq. (9b) is exponentially increasing, with an effective rate  $2\omega_s - \omega_M$ . As discussed above, this makes Eq. (9b) more and more self-consistent, with increasing time, even when applied to a *single*  $s$  mode.

#### APPENDIX B

We give in what follows some details about the passage from Eqs. (13), (14), and (15), to the self-consistent equation (21). Calculations are referred specifically to the CLM in three dimensions. First of all, we calculate the width (in frequency)  $\Delta\Omega(j)$  of the band of  $s$  modes around each quasisonant value given by Eq. (18a). From Eq. (16b) and

from the definition (17c), it is easy to see that

$$\Delta\Omega(j) = 2\xi/\tau \quad (\text{independent of } j). \quad (\text{B1})$$

The number  $M(j)$  of  $s$  modes around each quasisonant value  $k_j$  [Eq. (18a)], can be obtained by integrating the CLM density of elastic modes in a frequency interval of (small) width  $\Delta\Omega(j)$ , around each quasisonant frequency  $\Omega_0(k_j)$ . According to Eq. (B1), the result (to the lowest order in  $\xi$ ) is

$$M(j) = \xi \frac{V k_j^2}{\pi^2 c_0 \tau}. \quad (\text{B2})$$

From Eq. (14a), one gets for the CLM

$$\mathbf{q}_T^2(T_i) = \frac{3V\kappa T_i k_D}{2mN(\pi c_0)^2}, \quad (\text{B3a})$$

expressing the ergodic part of the square oscillation amplitude of each atom. To calculate the  $s$  part, we make the approximation that all the  $M(j)$   $s$  modes have the same rate of exponential increase  $\omega_s$ , given by Eq. (18b). From Eq. (14b), with the aid of Eq. (9c), it follows that

$$\mathbf{q}_s^2(T_i, t) = \frac{3\kappa T_i}{2mN} \sum_{j=0}^{j_M} \frac{M(j)}{\Omega_0(j)} \left[ \frac{e^{[2\omega_s - \omega_1(k_j)]t}}{\alpha} - 1 \right], \quad (\text{B3b})$$

where  $\omega_1(k_j)$  is given by the first term in Eq. (19) (for sufficiently low temperatures), and  $j_M$  is the  $j$  value corresponding to  $k_M(t)$  [Eq. (20c)]. Passing to continuous  $j$  values, the sum in Eq. (B3b) can be transformed into an integral in  $k$  space, since, from Eq. (18a),  $dk = (2\pi/\tau c_0) dj$ . Recalling Eq. (B3a) and (B2), expression (B3b) can be finally cast in the form

$$\mathbf{q}_s^2 = \mathbf{q}_T^2 \frac{\xi}{2\pi k_0} \left[ \int_{k_0}^{k_M} \frac{e^{[2\omega_s - \omega_1(k)]t}}{\alpha} dk - k_M + k_0 \right]. \quad (\text{B4})$$

From Eq. (B4), it is not difficult to get Eqs. (21), according to the procedure indicated in the text. In order to approach the solution of Eq. (21a), it is convenient to set  $z^2 = T/T_\infty$  and verify, with the aid of Eqs. (23a), (23b), and (21b), that  $F(T) = z$  in any case. Equation (21a) can be thereby put in a more convenient form:

$$z^2(x) = \frac{T_i}{T_\infty} \left[ 1 + \frac{\Gamma}{z} \left( \frac{e^{x^2}}{f(x)} \int_{zf(x)}^x e^{-y^2} dy + z - 1 \right) \right], \quad (\text{B5})$$

where  $\Gamma = \xi/\omega_D \tau$  for  $\tau < L/c_0$  and  $\Gamma = \xi a/L$  for  $\tau > L/c_0$ . This shows that the only parameters to be defined for a numerical solution of Eq. (21a) (Fig. 3), are just the ratio  $T_i/T_\infty$  and  $\Gamma$ .

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