ON THE DIRECTIVITY OF THREE-PHONON INTERACTIONS

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The quantum-mechanical conceptions of the phonon gas are applied for analysis of the three-phonon interaction. It is shown that the directivity of the processes in this interaction depends on the occupation numbers of the phonon branches and is determined by the tendency of the system to maximum entropy. At equilibrium, the energy is, in the general case, distributed nonuniformly among the branches, indicating that three-phonon interactions are insufficient to establish complete thermodynamic equilibrium. Cases of "coalescence" and decay of phonons are analyzed.

The invariance of the equations of hydrodynamics and elasticity theory without dissipation and dispersion with respect to the sign of the time and velocity (or the sign of the wave vector) implies reversibility of the wave processes, including nonlinear ones. Obviously, if the energy and quasi-momentum conservation conditions (synchronism condition) are satisfied in the forward three-phonon process (Fig. 1a) of "coalescence" of phonons in the third branch* $\Phi_1+\Phi_2\to\Phi_1'+\Phi_2'+\Phi_3'$ two decay processes $\Phi_1'+\Phi_3'\to\Phi_2;\Phi_2'+\Phi_3'\to\Phi_1$ and one "coalescence" process $\Phi_2'+\Phi_1'\to\Phi_3$, for each of which the synchronism conditions are also satisfied can be distinguished in the reverse process (Fig. 1b). It is easily seen that on the appearance of a "coalescence" branch in any three-phonon interaction, the possibility of the reverse decay process arises together with the forward process. At the same time, numerous experimental results indicate that nonlinear wave processes have a certain directivity: in some cases, phonon "coalescence" prevails, and in others the decay processes are dominant. Directivity of the nonlinear process is characteristic not only for coherent, strictly deterministic waves, where constancy of the relative phases of the interacting waves is of essential importance, but also for the cases in which the relative phases can vary randomly [1,2]. In the latter case, the directivity of the nonlinear wave process is determined by the conditions under which the ensemble of random waves tends to equilibrium. In this case, it is convenient to use the methods of nonequilibrium thermodynamics to determine the direction of the process, and this is done in the present paper.

Let us consider the three-phonon "coalescence" indicated in Fig. 1a. The total number of phonons before the interaction is $N = N_1 + N_2$, where N_1 and N_2 are the numbers of phonons in the first and second branches. The entropy before the interaction (see, for example, [3]) is

$$S_0 = \sum_{i=1}^{2} G_i\sigma(n_i), \tag{1}$$

where $\sigma(n_i)=(1+n_i)\ln(1+n_i)-n_i\ln n_i$, $n_i=N_i/G_i$ are the initial occupation numbers and G_i is the number of possible states in the phase space for phonons of the i-th branch. In the course of the interaction, the total number of phonons and their distribution among all of the three branches change. If N_1' and N_2' are the numbers of phonons that have not yet participated in the interaction, the present total number of phonons will be $N'=N_1'+N_2'+N_3'$, where $N_3'=N_i-N_i'$ $(i=1, 2)$. The entropy in the intermediate state is

$$S = \sum_{i=1}^{3} G_i'\sigma(n_i'), \tag{2}$$

*The term "branch" is used for phonons of approximately equal energy and momentum; these phonons may be in various states in phase space.

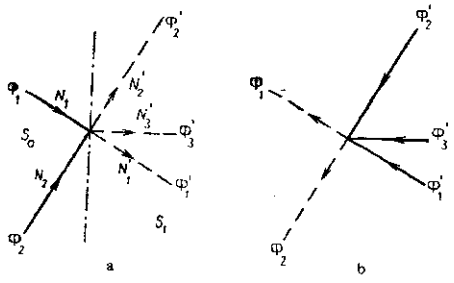

Fig. 1

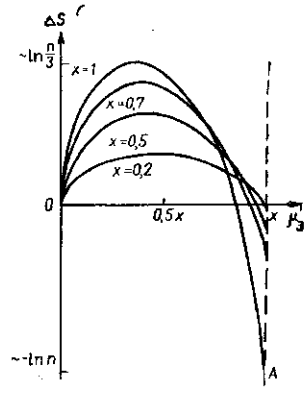

Fig. 2

where $n_i' = N_i'/G_i'$ are the present occupation numbers, which are related to the n_i by the obvious relation

$$n_i' = n_i g_i - \in \eta_i \quad (i \neq 3). \tag{3}$$

Here $g_i = G_i/G_i'$, $\in = N_3'/N$, $\eta_i = N/G_i'$. It is easily seen from (3) that when $\omega_3 = \omega_1 + \omega_2$, the initial energy $E_0 = \hbar \sum_{i=1}^{2} \omega_i G_i n_i$ is conserved in the interaction.

The entropy change

$$\Delta S = S - S_0 = \sum_{i=1}^{2} [G_i' \sigma (n_i g_i - \in \eta_i) - G_i \sigma (n_i)] + G_3' \sigma (\in \eta_3). \tag{4}$$

From (4)

$$\varkappa = \frac{\partial (\Delta S)}{\partial \in} = N \ln Y, \tag{5}$$

where

$$Y = \frac{1 + \in \eta_3}{\in \eta_3} \prod_{i=1}^{2} \frac{n_i g_i - \in \eta_i}{1 + n_i g_i - \in \eta_i}.$$

When the numbers n_i (i = 1, 2) are specified, equilibrium is established under the condition Y = 1, from which follows a quadratic equation for $\in$:

$$\begin{aligned} \in^2 (\eta_1 \eta_2 + \eta_1 \eta_3 + \eta_2 \eta_3) - \in [\eta_3 + n_1 g_1 (\eta_2 + \eta_3) + \\ + n_2 g_2 (\eta_1 + \eta_3)] + n_1 n_2 g_1 g_2 = 0. \end{aligned} \tag{6}$$

If it is assumed that the interaction does not change the number of possible states of the initial branches in phase space, $G_i = G_i'$ (i = 1, 2), transition to the present occupation numbers in accordance with (3) reduces (6) to a first-order equation:

$$n_{30} = \in \eta_{30} = \frac{n_{10} n_{20}}{1 + n_{10} + n_{20}}, \tag{7}$$

where the zero subscripts identify the equilibrium values of the occupation numbers. When the occupation numbers are small n_{i0} (i = 1, 2) it follows from (7) that the occupation number of the third branch is proportional to the product of the equilibrium occupation-number values of the first and second branches; when $n_{20} \simeq n_{10} = n_0 \gg 1$ we obtain $n_{30} \simeq n_0$. At large but sharply different occupation numbers $n_{20} \gg n_{10} \gg 1$, $n_{30} \simeq n_{10}$, i.e., the occupation number of the third branch is equal to the occupation number of the weak phonon branch. Hence follows the impossibility of "pumping" of all of the energy of the weak branch into the combination branch*, although under certain conditions the energy of the combination branch at equilibrium may exceed the energy of the weak branch.

*As we have already noted, the case of strict coherence is not considered here.

In (1)-(6), we did not sacrifice generality, assuming that each of the phonon branches has various regions G_i and that the dimensions of these regions change after the interaction, $G_i \neq G_i'$. Determination of these regions and their transformation constitute an independent problem that will not be considered in this paper. We can only say that not only "defocusing" in phase space, but, under certain conditions, also "focusing" which corresponds to "monochromatization" of the waves in time and space after the interaction, are evident possibilities in three-phonon interactions.

It will be interesting to express n_{30} in terms of the initial occupation numbers of the initial branches. We confine ourselves to consideration of the problem with no change in the numbers of states in phase space: $G_i = G_i'$ (i = 1, 2) and $G_1 = G_3 = G$; then $\varepsilon = 1$, $\eta_i = \eta = N/G$. It follows from the equilibrium condition (Y = 1) and (3) that $n_{30} < n_2$, n_1 always at equilibrium, i.e., it is not possible for all of the phonons (of at least one of the initial branches) to be transferred to the third branch. For convenience, we shall assume that $n_1 = nx$ and $n_2 = n$ ($x \leq 1$). We also introduce the relative occupation numbers $\mu_i' = n_i'/n$ (i = 1, 2, 3). The obvious relations

$$\mu_1' = x - \mu_3', \quad \mu_2' = 1 - \mu_3' \tag{8}$$

follow from (3). From (6), we have for the equilibrium occupation number of the third branch

$$n_{30} = \varepsilon\eta = \frac{1}{6}\{1 + 2n(1+x) \pm [1 + 4n^2(x^2 - x + 1) + 4n(x+1)]^{1/2}\}. \tag{9}$$

Let us consider the case $n \gg 1$. If we take the upper sign in (9), we obtain $\mu_3' \geq x$ for the root n_{30+} and condition (8) is not satisfied; $\mu_3' = x$ requires special treatment and the analysis shows that it is not an extreme point for ΔS. Therefore the root n_{30-} determines a unique stable equilibrium position with a maximum of ΔS.

The form of the function $\Delta S = \varphi(\mu_3')$ is shown in Fig. 2 for various x. At the beginning of the interaction as $\varepsilon \to 0$ and $\varkappa \sim \ln \frac{1}{\varepsilon} \to +\infty$ the entropy increases rapidly and active "coalescence" of phonons occurs. As the third branch is occupied, the reverse processes become increasingly important along with "coalescence" with the result that the entropy increases more slowly and, finally, equilibrium intervenes when the forward and reverse processes cancel one another. The efficiency of transition of phonons from the initial branches to the third branch depends on x and is highest at equal initial occupation numbers (x = 1). At equilibrium, the distribution of energy among all three branches for x = 1 will be uniform only in the particular case of equal frequencies ($\omega_1 = \omega_2$). If $x \neq 1$, an equilibrium energy distribution will be obtained only at a certain frequency ratio $\omega_2/\omega_1 = (x-1) + (x^2 - x + 1)^{1/2}$. This indicates that single three-phonon interactions are not sufficient in the general case to establish a uniform distribution of energy among all phonon branches and, consequently, that the Menli-Rou rule is limited.

It also follows from Fig. 2 that preferential "coalescence" of phonons occurs in the first and second branches at relative occupation numbers $\mu_3' < \mu_{30}$; the reverse decay processes prevail at $\mu_3' > \mu_{30}$. Consequently, the directivity of three-phonon processes is determined by the tendency of the system to equilibrium, which, in turn, depends on the occupation-number ratios. The irreversible nature of the three-phonon interactions in this statistical model is obvious. The ΔS diagram in Fig. 2 also yields qualitative information on the different types of three-phonon processes. For example, when $\gamma = x - \mu_3'$ is small but μ_2' is finite, the first branch is nearly unoccupied according to (8) and the entropy increase corresponds to the decay process $\Phi_2' + \Phi_3' \to \Phi_1'$; we note that here $\varkappa \sim \ln\gamma$ according to (5) and that as $\gamma \to 0$ $\varkappa \to -\infty$; hence follows activity at the beginning of the decay process. The rate of subsequent development of decay depends on $\mu_2' \simeq 1 - x = \delta$. At $\delta \ll 1$, the strong third-branch signal decays under the action of the weak second-branch signal; here the entropy increase corresponds to parametric amplification of the weak signal by a powerful pump μ whose frequency is higher than that of the signal.* At $\delta < 1$ (excepting $\delta = 1$, since then the ΔS

*The absence of an amplification threshold in this process is explained by the features of the model chosen, i.e., by the absence of a thermal phonon reservoir, which would result in a finite lifetime of the phonons under consideration.

diagram contracts to a point: $\mu_3' = 0$, $\Delta S = 0$), situations are possible in which the decay process advances with a low-frequency wave μ_2' that is stronger than μ_3'. The ΔS diagram shows that parametric amplification proceeds more efficiently the weaker the signal μ_2'. In the decay region $\Delta S < 0$, therefore, decay situations are less probable than "coalescence" situations, and parametric situations are even less probable, since they correspond to larger deviations of the system from equilibrium.

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