Lecture 8.

Kinetics of cascade processes. An example: ‘ballistic‘ phonons in dielectrics. Non-local phonon thermo-conductivity.

There are many phenomena in kinetics of the phonon systems in dielectrics. We present here one example of it.

Experiments with hot acoustic phonons. Elementary acts and their rates.

Consider the following experimental setup: a laser beam illuminates a crystal of $GaAs$ and, through light absorption, generates electron-hole pairs. These pairs recombine emitting optical phonons with nearly zero momentum $\mathbf{q} \approx 0$ and frequency $\omega_{LO}(0)$. Since the group velocity of optical phonons is very low, the excitation energy remains localized at the spots which the laser beam is focused at. Due to anharmonicity, the optical phonons (LO) decay into a pair of acoustic phonons A and A' with finite momenta $\mathbf{q}$ and $-\mathbf{q}$, which can be found from the energy-momentum conservation condition:

$$\omega_{LO}(0) = \omega_A(\mathbf{q}) + \omega'_A(-\mathbf{q})$$

Acoustic phonons are propagate through a crystal at the speed of sound ($\omega_A(q) = sq$), colliding with point defects[1] and expe-

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[1] Even in a very clean and carefully grown crystal, the nuclear masses fluctuate from site to site due to large number of isotopes of both Ga and As. These random fluctuations of the masses cause irregularities, which lead to elastic scattering of phonons.

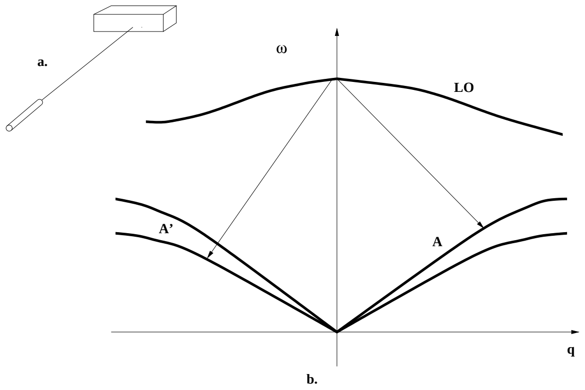

Figure 1: The scheme of a hot phonon experiment: a. The real space setup; b. Decay of an optical phonon into two acoustic

riencing anharmonic processes of a single phonon splitting into two and two phonons coagulating into one[2]. These processes lead to fast evolution, both in space and time, of the distribution function of acoustic phonons in their frequencies. Attaching local thermometers (bolometers) to the crystal, experimentalists measure the rate of energy propagation. The purpose of this lecture is to develop the relevant theory.

The simplest elementary process is elastic scattering of a phonon of frequency ω and wave vector $q = \omega/s$, say, by fluctuations of the uniform distribution of isotopes. Since the isotope irregularities in different elementary cells are not correlated, the scattering is equivalent to that by an impurity of atomic size a. The probability $w(\mathbf{q}, \mathbf{q}')$ of such a scattering per unit time is proportional to the speed of sound s multiplied by concentration of

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[2] The dominant process is the decay in which the phonon frequency decreases. Being repeated, this process causes a cascade, which has given the name to the whole phenomenon we are discussing in this lecture

imperfections $N \propto a^{-3}$ and by the scattering cross-section σ

$$w_e(\mathbf{q}, \mathbf{q}') = Nsd\sigma.$$

The scattering of a long wave-length phonon by a point-like scatterer obeys the Rayleigh law $\sigma \propto a^2(\omega/\omega_D)^4$ and depends on the second moment of the isotope distribution

$$\delta = \frac{\langle(\delta M_{Ga})^2\rangle + \langle(\delta M_{As})^2\rangle}{(M_{Ga} + M_{As})^2}.$$

Finally

$$w_e \sim \delta \, \frac{s}{a} \, \left(\frac{\omega}{\omega_D}\right)^4 . \qquad (1)$$

In the leading order in anharmonicity, there are the following inelastic processes

- spontaneous decay of a single phonon into two phonons;
- background induced decay;
- spontaneous coalescence;
- inelastic scattering by the background.

The probabilities of the processes are:

$$W_{1\to 2} = \sum_{\mathbf{q}_1\mathbf{q}_2} |M(q, q_1, q_2)|^2 N_q \left(1 + N_{q_1}\right) \left(1 + N_{q_2}\right) \delta_{\omega_{\mathbf{q}} - \omega_{\mathbf{q}_1} - \omega_{\mathbf{q}_2}}; \quad (2)$$

$$W_{2\to 1} = \sum_{\mathbf{q}_1\mathbf{q}_2} |M(q, q_1, q_2)|^2 \left(1 + N_{\mathbf{q}_1}\right) \left(1 + N_{\mathbf{q}_2}\right) N_{\mathbf{q}} \, \delta_{\omega_{\mathbf{q}_2} - \omega_{\mathbf{q}_1} - \omega_{\mathbf{q}}}. \quad (3)$$

The probability of the processes of spontaneous decay is given by Eq (2) in which the occupation numbers $N_{\mathbf{q}_{1,2}}$ are neglected. Keeping them in Eq (2) corresponds to taking into account the background induced decay processes. Equation (3) describes the probabilities of both spontaneous coalescence of phonons with momenta $\mathbf{q}$ and $\mathbf{q}_1$ (if $N_{\mathbf{q}_{1,2}}$ are neglected) and the background induced scattering of the $\mathbf{q}$-phonon by the $\mathbf{q}_1$-phonon with transformation into the $\mathbf{q}_2$-phonon. Under the condition of equilibrium and at $\omega_{\mathbf{q}} \approx T$, the occupation numbers are of the order of unity, being small at high-frequencies. In the course of the cascade processes, considered in this lecture, the occupation numbers remain small in a broad range of frequencies. Therefore, the cascades are dominated by the processes of spontaneous decay. Only at very low frequencies $\bar{\omega}$ the occupation numbers become of the order of unity. At these frequencies the rate of coalescence processes is comparable with that of decays, and, as a result, the distribution function at $\omega \sim \bar{\omega}$ has approximately Planckian form with the temperature $T \sim \bar{\omega}$. So, the complete frequency range $0 < \omega < \omega_* \approx \omega_{LO}/2$ can be divided into two intervals: the cascade interval $\bar{\omega} \gg \omega \leq \omega_*$, in which only processes of spontaneous decay are significant, and the range of thermolization $0 < \omega \leq \bar{\omega}$ in which the induced decay and coalescence processes are significant.

Boltzmann kinetics equation for hot phonons and the diffusion approximation

If we neglect the collisions, the phonon distribution function $N(t, \mathbf{r}, \mathbf{q})$ obeys the conservation law:

$$\frac{\partial N_{\mathbf{q}}}{\partial t} + \frac{\partial \omega(\mathbf{q})}{\partial \mathbf{q}} \frac{\partial N_{\mathbf{q}}}{\partial \mathbf{r}} = 0; \qquad \frac{\partial \omega(\mathbf{q})}{\partial \mathbf{q}} = s\,\frac{\mathbf{q}}{q}. \tag{4}$$

If the effect of collision is taken into account, a non-vanishing right-hand side appears in Eq(4), called “the collision integral”:

$$\frac{\partial N_{\mathbf{q}}}{\partial t} + s\,\frac{\mathbf{q}}{q}\,\frac{\partial N_{\mathbf{q}}}{\partial \mathbf{r}} = \mathrm{St}_e\{N_{\mathbf{q}}\} + \mathrm{St}_{pp}\{N_{\mathbf{q}}\} + G, \tag{5}$$

where St_e is responsible for elastic collisions, $\mathrm{St}_{pp}\{N_{\mathbf{q}}\}$ is responsible for the phonon-phonon collisions and $G \propto \delta\left[\omega(\mathbf{q}) - \omega_*\right]$ denotes a generation source, associated with the acoustic phonon pumping. This is the famous Boltzmann equation. Introducing new terms is, of course, a pretty innocent act. But, already, the assumption that these terms (in the equation for the distribution function $N(t, \mathbf{r}, \mathbf{q})$) depend only on the distribution functions taken at the same time t and the same point $\mathbf{r}$, is a conjecture[3], according to which neither two phonons nor a phonon and a scattering center, initially involved in the collision process, ever meet again. This presumption of a fast decay of correlations caused by the collision process needs, of course, a justification, which could be given and which we omit for the sake of a brevity.

The collision integral for the elastic collisions, St_e, expresses the probability of either scattering from the state $\mathbf{q}$ to the state $\mathbf{q}'$ or vice versa. If the probability of the elementary scattering act per unit time is $w(\mathbf{q}, \mathbf{q}') = w(\mathbf{q}', \mathbf{q})$, then the collision integral takes the form:

$$\begin{aligned}\mathrm{St}_e\{N_{\mathbf{q}}\} &= \sum_{\mathbf{q}'} w(\mathbf{q}', \mathbf{q}')N_{\mathbf{q}'}\left(1 + N_{\mathbf{q}}\right) - w(\mathbf{q}, \mathbf{q}')N_{\mathbf{q}}\left(1 + N_{\mathbf{q}'}\right) = \\ &= \sum_{\mathbf{q}'} w(\mathbf{q}', \mathbf{q}')\left(N_{\mathbf{q}'} - N_{\mathbf{q}}\right). \end{aligned} \tag{6}$$

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[3] The so called Boltzmannian “Stosszahlanzatz”

This gives the following expressions for the elastic relaxation rate τ_e^{-1} and the transport relaxation rate τ_{tr}^{-1}:

$$\frac{1}{\tau_e} = \sum_{\mathbf{q}'} w(\mathbf{q}', \mathbf{q}') \sim \frac{1}{\tau_{\mathrm{tr}}} = \sum_{\mathbf{q}'} w(\mathbf{q}', \mathbf{q}') \left[1 - \frac{\mathbf{q}\mathbf{q}'}{q^2}\right] \sim \delta \frac{s}{a} \left(\frac{\omega}{\omega_D}\right)^4 \tag{7}$$

The phonon-phonon collision integral $\mathrm{St}_{pp}\{N_{\mathbf{q}}\}$ describes the effect of phonon decays and coalescences on the distribution function. Using Eqs (2) and (3), we arrive at the expression:

$$\mathrm{St}_{pp}\{N_{\mathbf{q}}\} = S_{pp} = S_d + S_c; \tag{8}$$

$$S_d = \sum_{\mathbf{q}_1\mathbf{q}_2} \frac{|M|^2}{2} \{-N(1+N_1)(1+N_2) + N_1N_2(1+N)\}\, \delta_{\omega-\omega_1-\omega_2}; \tag{9}$$

$$S_c = \sum_{\mathbf{q}_1\mathbf{q}_2} |M|^2 \{-N_1N(1+N_2) + N_1(1+N_2)(1+N)\}\, \delta_{\omega_2-\omega_1-\omega} \tag{10}$$

Since the matrix element $M_{q\to 1+2}$ contains the wave vectors of incoming and outgoing phonons, it vanishes at small frequencies:

$$|M|^2 \propto \omega_1\omega_2\omega. \tag{11}$$

One can neglect $N_{1,2}$ in the cascade range of frequencies. As a result, the following estimate for the rate of *pp*-processes might be obtained:

$$S_d(\omega) \sim \sum |M|^2 \delta_{\omega-\omega_1-\omega_2} \sim |M|^2 g(\omega) \propto \omega^5. \tag{12}$$

Comparing this rate with the elastic one, given by Eq (7), we can see that in the time interval between two consecutive inelastic collisions there occur many elastic ones. This means that the momentum distribution relaxes much faster, than the distribution in frequencies and, therefore, the distribution function differs from the isotropic one by a small amount:

$$N(\mathbf{q}) = \bar{N}_q + \delta N(\mathbf{q}); \qquad \delta N \ll \bar{N}. \tag{13}$$

Using this inequality, and omitting, for the time being, the *pp*-collision integral, we can reduce the kinetic description to a diffusion one. For this purposes, we calculate the zero and the first moments of the Boltzmann equation with respect to the angular variable (the direction $\mathbf{q}/q$ of the wave vector):

$$\frac{\partial \bar{N}}{\partial t} + \frac{s}{q}\langle \mathbf{q}\nabla \delta N\rangle = S(\omega) + G(\omega); \tag{14}$$

$$\delta N = -\frac{s\tau_{\rm tr}}{q}\mathbf{q}\nabla \bar{N}. \tag{15}$$

As a result, we arrive at the diffusion equation:

$$\frac{\partial \bar{N}}{\partial t} - D(\omega)\nabla^2 \bar{N} = S(\omega) + G(\omega), \quad D(\omega) = \frac{s^2\tau_{\rm tr}(\omega)}{3}, \tag{16}$$

where $G(\omega)$ is the generation term and the decay collision integral $S(\omega)$ contains only $\bar{N}(\omega)$ and $\bar{N}(\omega_1)$:

$$S(\omega) = \omega^{-6}\int_{\omega}^{\omega_*} d\omega_1 \mathcal{W}\left(\frac{\omega_1}{\omega}\right)\bar{N}(\omega_1) - \frac{\bar{N}(\omega)}{\tau_d(\omega)}; \tag{17}$$

$$\frac{1}{\tau_d(\omega)} = \omega^{-6}\int_0^{\omega} d\omega_1 \mathcal{W}\left(\frac{\omega_1}{\omega}\right). \tag{18}$$

Scaling solution. Different experimental set-ups and corresponding responses.

Equation (16) is a fairly complicated integro-differential equation for the function $\bar{N}(\omega, \mathbf{r}, t)$ [4] One simplification we have

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[4] In what follows we will omit the “bar” sign.

already made, eliminating the dependence of the distribution function N on the direction of the phonon wave vector $\mathbf{q}$. A further simplification comes from the fact that the kernel $\mathcal{W}(x)$ of the integral equations (17) and (18) does not depend on the variables of integration ω_1 and ω_2 separately but only on their ratio $\omega_{1,2}/\omega$. Indeed, if $\omega \ll \omega_*$, then the limit $\omega_* \to \infty$ must be taken in Eq (17) and the relative frequency $x = \omega_1/\omega$ introduced, which leads to the collision integral (17) in the following form :

$$S(\omega) = \omega^{-5}\left\{\int_1^\infty \mathcal{W}(x)N(\omega x)\, dx - N(\omega)\int_0^1 \mathcal{W}(x)\, dx\right\}. \quad (19)$$

This form of the collision integral allows to introduce the following scaling:

$$\begin{aligned} &t = \eta\tau_d(\omega), \qquad \mathbf{r} = \mathbf{R}L_d(\omega), \\ &L_d(\omega) = \sqrt{D(\omega)\tau_d(\omega)} \sim \delta^{1/2} a \left(\frac{\omega_D}{\omega}\right)^{9/2}, \end{aligned} \quad (20)$$

so that

$$N(\omega; \mathbf{r}, t) = f(\omega, \eta; \mathbf{R}),$$

and

$$\frac{\partial f}{\partial \eta} - \frac{\partial^2 f}{\partial \mathbf{R}^2} = \int_1^\infty \mathcal{W}(x) f(\omega x)\, dx - f(\omega)\int_0^1 \mathcal{W}(x)\, dx + G. \quad (21)$$

We consider now several related problems associated with different experimental setups.

Spectral evolution in a homogeneously excited crystal.

Consider an experiment, in which the phonon generation is carried out uniformly over the sample volume by means of a short pulse at time $t = 0$ and frequency $\omega_* - \delta\omega \leq \omega \leq \omega_* + \delta\omega$. The total energy of the pulse is E, which means that the occupation number of initially generated phonons is $N_* = E/(\hbar\omega_*\,\delta\omega g(\omega_*))$, where $g(\omega)$ is the phonon density of states. At subsequent time $t \gg \tau_d(\omega_*)$ the distribution function $f(\omega)$ looks like it is shown in Fig 2.: a maximum at $\omega = \omega_t$ and a long tail towards high frequencies. Since the rate of decay $\tau_d(\omega)$ rapidly increases with decreasing frequency, the total time of the cascade t is of the order of the time of its last stage, $\tau_d(\omega_t)$

$$t \sim \tau_d(\omega_t) \quad \rightarrow \quad \omega_t \sim \omega_* \left[\frac{\tau_d(\omega_*)}{t}\right]^{1/5} \tag{22}$$

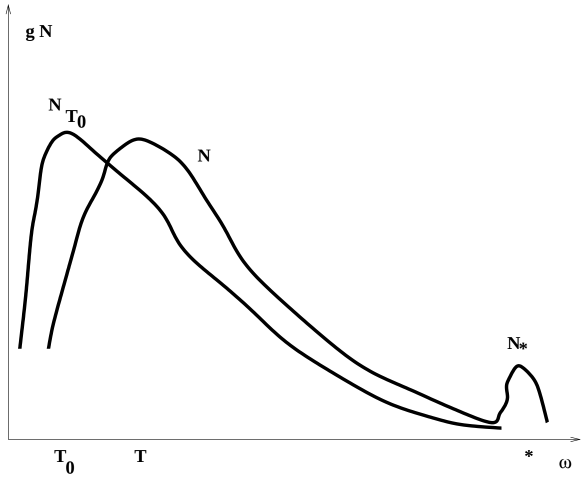

Figure 2: Time evolution of the spectrum for uniformly excited phonon system

Since the total energy is conserved, the spectrum $f(\omega)$ in the interval $\omega_t \ll \omega \ll \omega_*$ is determined by the the energy conservation condition:

$$\hbar\omega_* g(\omega_*)\delta\omega N_* = \int_{\omega_t}^{\infty} \hbar\omega g(\omega) f(\omega,t) d\omega. \tag{23}$$

Assuming a power-law solution

$$f(\omega,t) = f(\omega_t,t)\Phi\left(\frac{\omega}{\omega_t}\right); \quad \Phi(x) = \begin{cases} 1, & x = 1; \\ x^{\sigma}, & x \gg 1. \end{cases}$$

we, finally, deduce from Eqs (22) and (23)

$$f(\omega,t) = N_* \frac{\delta\omega}{\omega_*}\left(\frac{t}{\tau_d(\omega_*)}\right)^{4/5} \Phi\left(\frac{\omega}{\omega_t}\right). \tag{24}$$

where the function $\Phi(x)$ has the following asymptotes:

$$\Phi(x) = \begin{cases} 1, & x = 1; \\ x^{-4}, & x \gg 1. \end{cases} \tag{25}$$

A similar conclusion can be obtained directly from Eq(21). The energy conservation condition leads to the following equation:

$$\frac{\omega}{\tau_d(\omega)} = \omega^6 \int_0^1 dx x \mathcal{W}(x). \tag{26}$$

If we look for a power-law solution of Eq (21)

$$f(\omega,\eta) = A\omega^{\sigma}\phi(\eta),$$

then the function $\phi(\eta)$ obeys the equation:

$$\frac{\partial\phi(\eta)}{\partial\eta} = -\phi(\eta) + \int_0^1 dx\; x^{-\sigma-6} h(x)\phi\left(\frac{\eta}{x^5}\right);$$
$$h(x) = \frac{\mathcal{W}(x)}{\int_0^1 y^5 \mathcal{W}(y) dy}. \tag{27}$$

The index σ can be found from the energy conservation. In the long time limit $\eta \gg 1$ the total energy E(t) is equal to:

$$
\begin{aligned}
E(t) &= \int_0^{\omega} d\omega g(\omega)\hbar\omega f(\omega,t) = \\
&= \frac{A}{5}[\omega^{\sigma+2}\ g(\omega)]_{\omega=\omega_*} \int_0^{\eta_*} \frac{d\eta}{\eta}\left(\frac{\eta}{\eta_*}\right)^{(\sigma+4)/5} \phi(\eta) = E_*. \quad (28)
\end{aligned}
$$

with $\eta_* = t\tau_d^{-1}(\omega_*) \gg 1$. The integral on the right-hand side of Eq(28) does not depend on η_* if $\sigma = -4$. Under this condition, the normalization reads

$$
\frac{1}{5}\int_0^{\infty} \frac{d\eta}{\eta}\phi(\eta) = 1, \qquad A = \frac{2\pi^2 s^3}{3}\ E_*. \quad (29)
$$

The dimensionless equation for the dimensionless function $\phi(\eta)$

$$
\begin{aligned}
\frac{\partial\phi(\eta)}{\partial\eta} &= -\phi(\eta) + \int_0^1 dx h(x)\phi\left(\frac{\eta}{x^5}\right); \\
h(x) &= \frac{\mathcal{W}(x)}{\int_0^1 y^5 \mathcal{W}(y) dy} \quad (30)
\end{aligned}
$$

should still be solved, which could be done, using the Mellin transform.

Finally, the time of thermolization τ_T could be found from the estimate

$$
\tau_T = \tau_d(\omega = T) \sim \tau_d(\omega_*)\ \left(\frac{\omega_*}{T}\right)^5.
$$

Stationary excitation.

Consider a steady and uniform excitation at $\omega = \omega_*$. Under this condition the flux $Q = E(\omega)\tau_d(\omega)$ down in frequency does not

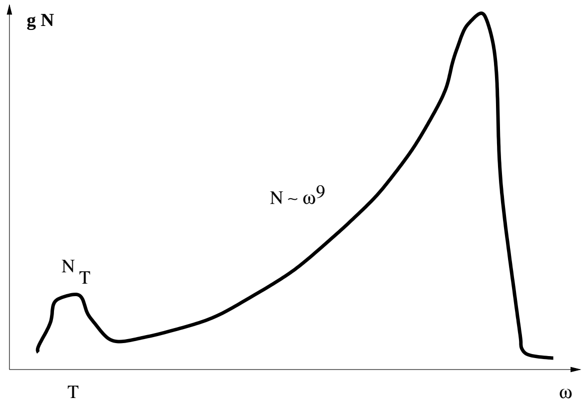

Figure 3: Phonon spectral density in the case of stationary excitation

depend on frequency ω. Equation (21) takes the form

$$S(\omega) + G(\omega); \qquad G(\omega) = \frac{Q}{g(\omega_*)\hbar\omega_*}\,\delta(\omega - \omega_*), \tag{31}$$

which leads to the integral equation

$$\int_0^1 h(x)\; x^{\sigma-5}\frac{dx}{x} \tag{32}$$

with the normalization condition

$$\int_0^1 h(x)\; x^3\; dx = 1. \tag{33}$$

Equations (32) and (33) lead jointly to the conclusion that $\sigma = -9$. The distribution function $f(\omega)$ has a power-law tail at high frequencies:

$$f(\omega) = \frac{Q\tau_d(\omega)}{\hbar\omega(\omega/s)^3} \propto \omega^{-9}.$$

The spectrum is shown in Fig 3.

Local excitation and signal arrival time.

Since the down-conversion occurs slower than the elastic scattering, the initial hot spot will spread out diffusively and in time $\tau_d(\omega)$ cover an area of radius $L(\omega) = \sqrt{D(\omega)\tau_d(\omega)} \sim s\sqrt{\tau_e(\omega)\tau_d(\omega)} \propto \omega^{-9/2}$. On the other hand, since the diffusion coefficient $D(\omega)$ strongly increases with the frequency down-conversion, the contribution of the last stage of conversion dominates the spread. Therefore, the characteristic frequency ω_t at the time t is determined by Eq (22) and the arrival time t_L at the point situated at a distance L is determined from

$$L = L(\omega_t) \propto \omega_t^{-9/2} \propto t^{9/10}, \quad \to \quad t_L \propto L^{10/9}.$$

Therefore, the down-conversion makes the diffusive spread look more like ballistic propagation.