# THE POSSIBILITY OF SECOND SOUND WAVES REGISTRATION IN ISOTOPIC ENRICHED DIAMOND SINGLE CRYSTAL

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Using the experimental data of diamond thermal conductivity we obtain the data of normal and resistive processes of phonon interaction in Callaway approach. As the result we obtain the concentration values of $^{13}C$ in diamond single crystal, under which the isotropic scattering is weak, and the temperature range of second sound waves existence in diamond.

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## 1. INTRODUCTION

Modern technologies give us opportunity to obtain isotopic pure perfect single crystals. It's been experimentally established that the thermal conductivity coefficient of such crystals has maximum in low temperature range [1]. Especially it becomes apparent in diamond crystals. This fact is caused by a special role played by normal processes (where the conservation law of quasi-impulse is present) in interaction between phonons. In perfect single crystals the propagation of weakly damping second sound waves, similar to HeII ones, is possible. The most probable at that are the normal processes of phonon interaction comparatively to resistive ones (where the conservation law of quasi-impulse fails). The major contribution to second sound waves damping makes isotopic phonon scattering. For the first time second sound waves were registered in NaF [2] single crystal and solid $^4$He [3], where the isotopic damping is absent at the temperature range of 10…20 K.

Using the data of normal and resistive processes of phonon interaction (umklapp processes, phonon scattering on impurities, isotopes and on bounds of sample), obtained while the description of experimental data of diamond thermal conductivity coefficient in Callaway model, the concentration values of $^{13}$C are obtained, under which the isotopic scattering is weak, the temperature range of second sound waves existence in diamond is established. Starting from $^{13}$C isotope concentration equal to 0.1 % and less the second sound waves registration is possible in the temperature range near maximum of thermal conductivity of diamond at T=104 K. This temperature is one order higher than the temperature of second sound waves registration in NaF and solid $^4$He.

## 2. EQUATIONS OF GAS DYNAMICS OF QUASI-PARTICLES WITH TAKING INTO ACCOUNT EXTERNAL FIELDS

In the kinetic theory the state of gas of quasi-particles is characterized by distribution function of quasi-particles $N \equiv N^j(\mathbf{p},\ \mathbf{r},\ t)$, which satisfies kinetic equation which has a form of Boltzmann equation:

$$\left(\frac{\partial}{\partial t}+\mathbf{g}\frac{\partial}{\partial \mathbf{r}}-\frac{\partial \varepsilon}{\partial \mathbf{r}}\frac{\partial}{\partial \mathbf{p}}\right)\ N=\left(\dot{N}\right)_{cm}, \tag{1}$$

where $\mathbf{g}\equiv\mathbf{g}^{(j)}=\partial\varepsilon^{(j)}/\partial\mathbf{p}$ is the group velocity of quasi-particles; $\varepsilon\equiv\varepsilon^{(j)}(\mathbf{p},\mathbf{r},t)$ is the Hamiltonian of quasi-particle, which is equal to its local energy; $\left(\dot{N}\right)_{cm}$ is the collision integral of quasi-particles, which takes into account the processes of collision, merge, fission and radiation of quasi-particles.

If we designate $\tau_N$ as a character time of phonon interaction concerned with *N*-processes (where phonon energy and quasi-impulse conservation laws are accomplished), and $\tau_R$ as a character time of resistive processes of phonon interaction (where these laws fail), the condition of predomination of normal processes is written as

$$\tau_N << \tau_R. \tag{2}$$

If at some moment of time the system of quasi-particles moves out of its equilibrium state, then in time $\tau_N$ quasi-local balance is established, which is characterized by distributive function $N_0^{(j)}$, which turns to zero the collision integral because of *N*-processes , and it is written as :

$$N_0^{(j)}=\left(\exp\frac{\varepsilon^{(j)}-(\mathbf{pu})}{T_0(1+\theta)}-1\right)^{-1}, \tag{3}$$

where $\mathbf{u}$ is drift velocity in gas of quasi-particles, $\Theta = (T - T_0)/T_0$ is relative temperature.

In the sate of gas of quasi-particles, which is close to local statistic balance, the solution of equation (1) in the gas dynamical approximation we search as:

$$N^{(j)} = N_0^{(j)} + \delta N^{(j)}, \ \left( \left| \delta N^{(j)} \right| << N_0^{(j)} \right), \tag{4}$$

where $N_0^{(j)}$ is the local equilibrium distributive function, which depends on gas dynamical quantities, and $\delta N^{(j)}$ depends on its gradients.

If we input thermodynamical potential $F_0$ as:

$$F_0 = -T \sum_{\mathbf{k},j} \ln\left(1 + N_0^{(j)}\right), \tag{5}$$

then knowing it as a function of $T$, $\mathbf{u}$, $\hat{A}_j$, which satisfies thermodynamical identity:

$$dF_0 = -S_0 dT - \mathbf{P} d\mathbf{u}, \tag{6}$$

we obtain impulse density $\mathbf{P}$, heat capacity $C$ and entropy $S$ densities, components of density tensor of quasi-particles $\tilde{\rho}_{ij}$:

$$\mathbf{P} = -\left(\frac{\partial F_0}{\partial \mathbf{u}}\right)_{T,\hat{A}}; \ S_0 = -\left(\frac{\partial F_0}{\partial T}\right)_{\mathbf{u},\hat{A}};$$

$$C = T\left(\frac{\partial S_0}{\partial T}\right)_{\mathbf{u},\hat{A}}; \ \bar{\tilde{\rho}}_{il} = \frac{\partial \bar{\bar{P}}_i}{\partial u_l} = -\left(\frac{\overline{\partial^2 F_0}}{\partial u_i \cdot \partial u_l}\right)_{T,\hat{A}}. \tag{7}$$

Applying the standard procedure [4] from kinetic equation (1) it is possible to obtain a system of gas dynamical equations, that describe an isotropic gas of quasi-particles in linear approximation on drift velocity $\mathbf{u}$ in reduced isotropic crystal model:

$$\dot{\mathbf{P}} + ST\nabla\theta = -r\mathbf{u} + \tilde{\eta}\Delta\mathbf{u} + \left(\tilde{\zeta} + \frac{\tilde{\eta}}{3}\right) \cdot \nabla div\mathbf{u};$$

$$C\dot{\theta} + Sdiv\mathbf{u} = \tilde{\kappa}\Delta\theta, \tag{8}$$

where $\mathbf{P} = \tilde{\rho} \cdot \mathbf{u}$, $\tilde{\rho}$ is the density of quasi-particles number, $r$ is the external friction coefficient, caused by processes of phonon interaction without quasi-impulse conservation (umklapp processes, phonon scattering on impurities, isotopes and sample boundaries), $\tilde{\eta}$, $\tilde{\zeta}$ are the first and second viscosity coefficients, $\tilde{\kappa}$ is the thermal conductivity coefficient.

## 3. PROBLEM SETTING AND ITS SOLUTION IN REDUCED ISOTROPIC CRYSTAL AND CALLAWAY MODELS

The secondary waves (SW) propagation in Bose gas of quasi-particles in solids was studied in work [4] basing on gas dynamics equations of quasi-particles, especially SSW in phonon gas in dielectric crystals.

The solution of equations (9) for all variables we'll search as $\exp\left[i\left(\Omega t - \tilde{\mathbf{k}}\mathbf{r}\right)\right]$ with frequency $\Omega$ and wave vector $\tilde{\mathbf{k}}$. From the compatibility condition of system of equations we obtain the dispersion equation for secondary waves (SW) [4]:

$$\Omega\left(\Omega^2 - W_{II}^2 \tilde{k}^2\right) - 2iW_{II}^2 \tilde{k}^2 \Gamma_{II} = 0. \tag{9}$$

From the dispersion equation (9) follows that $W_{II}$ is the phase velocity of SW and quantity $\Gamma_{II}$ is the damping coefficient of SW. Expressions for $W_{II}$ и $\Gamma_{II}$ have a simple structure for SSW phonon gas dynamics in reduced isotropic crystal model [4]:

$$W_{II} = \left(TS^2/C\tilde{\rho}\right)^{1/2};$$

$$\Gamma_{II} = \frac{r}{2\tilde{\rho}} + \left[\frac{1}{2\tilde{\rho}}\left(\frac{4}{3}\tilde{\eta} + \tilde{\zeta}\right) + \frac{1}{2C}\tilde{\kappa}\right]\tilde{k}^2, \tag{10}$$

where $C = 3S = \frac{3V_t^2}{(2+\beta^5)T}\tilde{\rho} = \frac{2\pi^2}{15}\frac{k_B^4 T^4}{V_t^3 \hbar^3}(2+\beta^3)$ is obtained in reduced isotropic crystal model in low temperature range. By low temperature range we mean the range of temperature where $\Theta_D/T >> 1$; $\Theta_D = 2\pi\hbar V_t/k_B a$ is the Debye temperature, $V_l, V_t$ are the velocities of transverse and longitudinal phonons, $\beta = V_t/V_l$, $a$ is the lattice parameter. Diamond single crystal has the highest Debye temperature ($\Theta_D \approx 1845K$) among other crystals, that's why the temperature in order of 100 K and lower can be considered as low for diamond.

If to introduce diffusive lengths of free path and related with them diffusive times by following expressions:

$$\tau_{\tilde{\eta}} = \frac{\tilde{\eta}}{\tilde{\rho}W_{II}^2}; \ \tau_{\tilde{\zeta}} = \frac{\tilde{\zeta}}{\tilde{\rho}W_{II}^2}; \ \tau_{\tilde{\kappa}} = \frac{\tilde{\kappa}}{CW_{II}^2}; \ \tau_R = \frac{\tilde{\rho}}{r}, \tag{11}$$

$\Gamma_{II}$ can be written in the following way:

$$\Gamma_{II} = \frac{1}{2}\left[\frac{1}{\tau_R} + \left(\frac{4}{3}\tau_{\tilde{\eta}} + \tau_{\tilde{\zeta}}\right)\Omega^2 + \tau_{\tilde{\kappa}}\Omega^2\right]. \tag{12}$$

The condition of weak damping SSW existence ($\Gamma_{II} << \Omega$) leads to next condition imposed on frequency $\Omega$, known as SSW existence "window" [4]:

$$\min\{\nu_{\tilde{\eta}}, \nu_{\tilde{\zeta}}, \nu_{\tilde{\kappa}}\} >> \Omega >> \nu_R, \tag{13}$$

where the collision frequencies $\nu_i$ are related to diffusive times $\tau_i$ by the expression $\nu_i = 1/\tau_i$.

The calculation of kinetic coefficients of phonon gas dynamics in the model of reduced isotropic crystal relatively to elasticity coefficient in [4] shows that basic role plays phonon viscosity. The number, similar to Prantl number in gas theory $\mathrm{Pr} = \frac{C\tilde{\eta}}{\tilde{\rho}\tilde{\kappa}}$, for phonon gas for most of crystals is equal to $10^2$ with a great degree of accuracy.

Further on we'll take into account that in reduced isotropic crystal model energy and quasi-impulse conservation laws allow $t + t \leftrightarrow l + g$ and $l + t \leftrightarrow l + g$ umklapp processes.

Using the Callaway [5] model for description of experimental data of thermal conductivity of diamond in low temperature range [6], we can calculate frequencies of phonon collisions, caused by normal and umklapp processes and also by scattering on isotopes and sample boundaries:

$$\nu_N = \frac{k_B A}{2\pi\hbar\rho a^6} T^4 5!\zeta(5);$$

$$\nu_{iso} = c_i \frac{{k_B}^4 a^3}{4\pi\hbar^4 {v_s}^3} T^4 \left(\frac{\Delta M}{M}\right)^2 8!\zeta(8); \qquad (14)$$

$$\nu_U = \frac{{k_B}^2 B}{(2\pi\hbar)^2 v_s} T^3 e^{-\frac{C}{T}} 6!\zeta(6);\ \nu_b = \frac{v_s}{D} 4!\zeta(4),$$

where $c_i = \frac{N_i}{N}$ is the isotope concentration, $\Delta M$ is the difference in isotope and the basic atom masses, $M$ is the mass of basic atom, $\zeta(n)$ is the Riemann zeta-function, $\rho$ is the diamond crystal density, $A = 7,2 \cdot 10^{-11} K^{-3}$, $B = 1,5 \cdot 10^{-12}$ cm/K, $C = 670K$, $D = 0,33$ cm are phenomenological constants obtained from experiment [6].

## 4. RESULTS ANALYSIS AND CONCLUSION

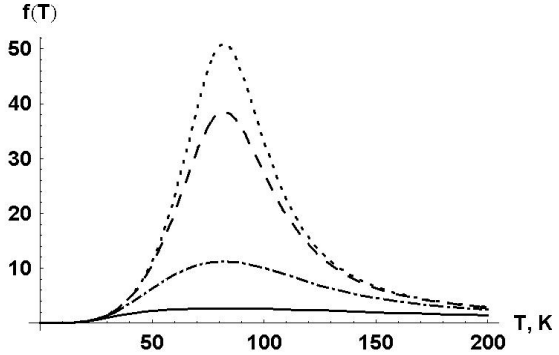
For making numerical estimations and obtaining the condition of weakly damping second sound waves existence in diamond single crystal we use the following data:

$\rho = 3,512$ g/cm$^2$, $\beta = 0,68$, $V_l = 1.81 \cdot 10^6$ cm/s,

$V_t = 1.235 \cdot 10^6$ cm/s, $W_{II} = 0.76 \cdot 10^6$ cm/s,

where $V_l$, $V_t$ and $W_{II}$ were calculated in reduced isotropic crystal model [4]. Using the Callaway model we examine inequality (13) to obtain the "window" of SSW existence or at least its low limit and the isotope concentrations (the main isotope is $^{13}C$, $\Delta M = 1$) of such "window". Substituting expressions (14) into inequality (13) we obtain SSW existence "window" in diamond single crystal. It's convenient to introduce function $f(T) = \nu_N / (\nu_U + \nu_{iso} + \nu_b)$, which value range $f(T) >> 1$ will provide the SSW existence "window". The following figure gives temperature dependence of function f(T) for different concentrations of $^{13}C$ isotope:

In the figure we can see that diamond has the SSW existence "window" in the region of $T \approx 80\ K$, starting from $c_i \approx 10^{-4}$ and less. Such form of curves is provided by competition of normal and resistive processes. In high temperature range (higher than $T \approx 80\ K$) umklapp processes predominate, in temperature range lower than $T \approx 80\ K$ normal processes and processes of phonon scattering on impurities and boundaries predominate.




*Solid line* – $c_i = 5 \cdot 10^{-4}$, *dash-dot line* – $c_i = 10^{-4}$, *dash line* – $c_i = 10^{-5}$, *dot line* – $c_i = 10^{-6}$

Thus the carried out analysis gives us reason to affirm that SSW in diamond crystal can be registered at temperatures in the range of $T \approx 100\ K$, if $^{13}C$ isotope concentration is lower than $10^{-4}$.

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**ВОЗМОЖНОСТЬ НАБЛЮДЕНИЯ ВОЛН ВТОРОГО ЗВУКА В ИЗОТОПИЧЕСКИ ОБОГАЩЁННОМ МОНОКРИСТАЛЛЕ АЛМАЗА**


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Используя экспериментальные данные по теплопроводности алмаза, в модели Калавэя получена информация о нормальных и резистивных процессах фононного рассеяния. Как следствие получены концентрации примеси изотопа $^{13}C$ в монокристалле алмаза, при которых изотопическое рассеяние фононов становится незначительным, а также промежуток температур, в котором существуют волны второго звука.


**МОЖЛИВІСТЬ СПОСТЕРЕЖЕННЯ ХВИЛЬ ДРУГОГО ЗВУКУ В ІЗОТОПІЧНО ЗБАГАЧЕНОМУ МОНОКРИСТАЛІ АЛМАЗУ**


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Використовуючи експериментальні дані теплопровідності алмазу, в моделі Калавея здобуто інформацію про нормальні та резистивні процеси фононного розсіяння. Як наслідок, здобуто величини концентрацій ізотопічних домішок $^{13}C$ в монокристалі алмазу, за яких ізотопічне розсіяння фононів стає незначним, а також проміжок температур, в якому існують хвилі другого звуку.