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On the Existence of Phonon Coherent States in Nanomaterials

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Abstract

In our work we describe the energy transfer by thermal excitations with accounting a coherence in nanosized systems during heat removal. A general formalism of thermal conductivity by second quantization method is proposed with an account of both the usual phonon model of heat transfer and the formation of coherent Schrödinger states of the oscillator system. An exact general form of solution for a time-dependent problem with arbitrary initial conditions is analytically developed. It is shown that at certain ratios of constants characterizing the interaction of phonons with the electronic subsystem a heat flow does not decay with time in the crystal.

Keywords: Coherent states; Coherent waves; Coherent heat flow; Nanocrystals; Thermal superconductivity

1. INTRODUCTION

Theoretical and experimental studies of recent years have shown that the transfer of thermal energy in dielectrics at the nano- and microlevel often has a quantum nature. It leads to new physical effects such as phonon coherence, thermal superconductivity, thermal echo, ballistic resonance nonlinear phenomena like solitons, etc. These phenomena can be used to control energy flows and to create fundamentally new devices. Besides the fundamental aspect of the problem, it will contribute to develop effective methods of heat removal for the design of nano/microelectronic devices and devices operating in space. The spatial coherence length and a temporal coherence length should simultaneously influence the thermal transport; their interplay is also critical for the engineering of nanoscale thermal transport. On the other hand, the dimensionality and system size have significant impacts on phonon coherence and on the coherent thermal transport.

Coherent thermal transport, including minimum thermal conductivity and Anderson localization, has been intensively observed in various nanophononic crystals. Nanophononic crystals are artificial materials and provide a new opportunity to engineer thermal properties of materials using the wave nature of phonons [1–3]. Phonon coherence has a significant impact on phonon-phonon scatterings, phonon modal correlations, and interfacial phonon propagation, which are different from the pure particle picture and also make coherence a critical attribute for phonons [4–6]. Because of the importance of coherent phonons in engineering thermal transport, the recent efforts to activate coherent phonon in real materials are increasing. Such attempts were done via a laser beam [7,8], or via the natural thermal fluctuations [9], which was previously studied for acoustic phonons at gigahertz frequency in optomechanical systems. In the paper [9], a general heat conduction theory is proposed to establish an original expression for the thermal conductivity that includes the full coherent nature

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of phonon excitations. This expression involves both phonon lifetimes and coherence times. It was supposed the particle-like behavior predominatly follows an exponential decay law $\mathrm{Cor}_\lambda(t) = \exp(-t/\tau_\lambda^p)$ with the lifetime τ_λ^p according to the conventional single-mode relaxation time (SMRT) approximation [10]. Besides, it was supposed that a correction should be considered, as the coherence effect increases, in the description of the phonon decay by including the modal coherence time τ_λ^c as follows $\mathrm{Cor}_\lambda(t) = \exp[-t/(2\tau_\lambda^p)]\exp[-4\ln(2t^2/(\tau_\lambda^c)^2)]$. The second Gaussian term originates from the interference between different modes, expressing coherence effects in the phonon dynamics.

Semiconductor optomechanics based on the use of exciton polaritons, is rapidly developed field due to advances in technologies for creating planar nanosystems. Polaritons are hybrid quasiparticles that are a mixture of photons and material excitations. Recently, polariton-pumped phonon lasers and dynamically tunable arrays of polariton parametric cavities have been implemented. A feature of the polariton is the presence of a resonant photoelastic interaction in addition to the geometric optomechanical interaction. Both exciton and photon components of the polariton can interact with an acoustic wave, which can lead to an increase in the optomechanical interaction. Theoretical study of optically-pumped excitons interaction with acoustic waves in planar semiconductor nanostructures in the strongly nonlinear regime was done in the work [11]. The results are applied to nonlinear sound propagation in the arrays of quantum wells or in the plane of Bragg semiconductor microcavities hosting excitonic polaritons.

In our work we will show that in a crystal, taking into account the interaction of vibrations of the crystal lattice with the electronic subsystem, coherent states similar to Schrödinger states can be formed. Taking into account the constant of nonlinear optomechanical interaction between optically pumped excitons and propagating acoustic waves, calculated in Ref. [11], we will show that coherent states during thermal transport are actually formed into a wave packet, and, in coordinate representation, are nothing more than an oscillator with a displaced center. These coherent states, with a Poisson distribution of the number of phonons in each mode, play an important role and lead to characteristic time dependences of heat fluxes. At certain phonon decay times in the frequency band associated with this constant, a heat flux (spectral flux density in the frequency band) that does not decay with time can be formed. This behavior of the heat flow is reminiscent of superthermal conductivity, considered in Refs. [12–17]. Semiconductors, cubic boron arsenide crystals (BAs), have recently been discovered [12–15] to have ultra-high thermal conductivity compared to most conventional materials and show great promise for thermal management in electronics [15–17].

2. PHONON COHERENT STATES

Elastic vibrations of the crystal lattice are considered as a set of plane waves, and in the quantum description — as a set of $n_{\mathbf{k}\lambda}$ phonons with different momenta $\mathbf{p} = \hbar\mathbf{k}$ ($\mathbf{k}$ is the wave vector), being in polarization states λ ($\lambda = 0,1,2$) and having frequencies $\nu = ck/2\pi = \omega/2\pi$ where ω is an angular frequency. The number of phonons of each type will be denoted as $n_{\mathbf{k}\lambda}$. We consider an influence of electrons on the phonon subsystem. We will use a notation $|\eta_i\rangle$ for the electronic state of the system, and $|i\rangle = |n_{\mathbf{k}\lambda}^{(i)}\eta_i\rangle$ for the initial "electronic & phonons" state of the system. Under the influence of phonons, described by the interaction of electrons with phonons $\hat{V}_{eph}$, the electronic system goes over to the state f (or $|f\rangle = |n_{\mathbf{k}\lambda}^{(f)}\eta_f\rangle$). In this state it can be described by a set of quantum numbers η_f of the material matter, and phonons, characterized by a set number of phonons with the same wave vectors and in the same polarization states, $n_{\mathbf{k}\lambda}^{(f)}$. As a result of the transition, one or more phonons are emitted or absorbed, in some polarization states with different pulses and frequencies. Differences in *momenta and polarization states* for phonons initiating the transition and for phonons emitted/absorbed as a result of the transition determine the polarization characteristics of the emitted or absorbed heat radiation. They are described *by differential transition probabilities,* $dw_{fi}^{(a)}$ (for absorption) and $dw_{fi}^{(e)}$ (for emission). We will pay considerable attention to the discussion of these quantities.

As is well known (and will be discussed below), the interaction of electrons with a deformation field can be considered by methods of perturbation theory. According to the general formulae of the theory of time-dependent perturbations, the probability $P_{fi}(t)$ of transition from state $|i\rangle$ to state $|f\rangle$ under the influence of a perturbation that acts in the time interval $[0,t]$ can be represented in the form

$$P_{fi}(t) = \left|\langle f|\hat{\mathcal{W}}^{(1)}(t)|i\rangle + \langle f|\hat{\mathcal{W}}^{(2)}(t)|i\rangle + \ldots\right|^2.$$

Here the transition operators $\hat{\mathcal{W}}^{(1)}$, $\hat{\mathcal{W}}^{(2)}$, $\hat{\mathcal{W}}^{(3)}$ corresponding to the harmonic perturbation can be represented as for one-quantum, two-quantum and so on transitions. For one-quantum $\hat{\mathcal{W}}^{(1)}$ (one-phonon) transitions accompanied by

emission (*e*) or absorption (*a*) of one phonon, characterized by quantities $\pi_\rho \equiv \{\mathbf{k}, \lambda, \omega_k\}$ whose probabilities will be determined mainly using the following of formulas

$$\langle f|\hat{w}^{(1)}(t)|i\rangle = -\frac{i}{\hbar}\int_0^t \langle f|\hat{V}(t')|i\rangle dt',$$

we have

$$\hat{V}(t') = \hat{V}^{(e)}(\pi_p)e^{i\omega_k t'} + \hat{V}^{(a)}(\pi_p)e^{-i\omega_k t'},$$

where

$$\hat{V}^{(e)}(\pi_p) = -\gamma\sqrt{\frac{\hbar}{2m\omega_k}}\hat{b}^\dagger_{\mathbf{k}\lambda}(0)\left(\hat{\boldsymbol{\alpha}}\cdot\mathscr{C}^*_{\mathbf{k}\lambda}(\mathbf{r})\right),$$

$$\hat{V}^{(a)}(\pi_p) = -\gamma\sqrt{\frac{\hbar}{2m\omega_k}}\hat{b}_{\mathbf{k}\lambda}(0)\left(\hat{\boldsymbol{\alpha}}\cdot\mathscr{C}_{\mathbf{k}\lambda}(\mathbf{r})\right). \quad (1)$$

Here

$$\hat{b}^\dagger_{\mathbf{k}\lambda} = \sqrt{\frac{\omega_k}{2\hbar}}\left(\hat{Q}_{\mathbf{k}\lambda} + \frac{\hat{P}_{\mathbf{k}\lambda}}{i\omega_k}\right),\quad \hat{b}_{\mathbf{k}\lambda} = \sqrt{\frac{\omega_k}{2\hbar}}\left(\hat{Q}_{\mathbf{k}\lambda} - \frac{\hat{P}_{\mathbf{k}\lambda}}{i\omega_k}\right) \quad (2)$$

are the operators of creation $\hat{b}^\dagger_{\mathbf{k}\lambda}$ and annihilation $\hat{b}_{\mathbf{k}\lambda}$ of the phonon in the state with the wave vector **k** and polarization λ, $\left[\hat{b}_{\mathbf{k}'\lambda'}, \hat{b}^\dagger_{\mathbf{k}\lambda}\right] = \delta_{\mathbf{k}'\mathbf{k}}\delta_{\lambda'\lambda}$, $\mathscr{C}_{\mathbf{k}\lambda}(\mathbf{r})$ is a coordinate part of the vector displacement operator $\hat{\boldsymbol{\xi}}_n(\mathbf{r},t)$ of the *n*th site of the crystal lattice:

$$\mathscr{C}_{\mathbf{k}\lambda}(\mathbf{r}) = \frac{1}{\sqrt{N}}\mathbf{e}_{k\lambda}e^{i\mathbf{k}\cdot\mathbf{r}},$$

$$\hat{\boldsymbol{\xi}}_n(\mathbf{r},t) = \sum_{\mathbf{k},\lambda}\sqrt{\frac{\hbar}{2m\omega_k}}\left[\hat{b}_{k\lambda}e^{-i\omega_k t}\mathscr{C}_{\mathbf{k}\lambda}(\mathbf{r}) + \hat{b}^\dagger_{k\lambda}e^{i\omega_k t}\mathscr{C}^*_{\mathbf{k}\lambda}(\mathbf{r})\right] =$$

$$= \sum_{\mathbf{k},\lambda}\sqrt{\frac{\hbar}{2m\omega_k}}\left[\hat{b}_{k\lambda}(t)\mathscr{C}_{\mathbf{k}\lambda}(\mathbf{r}) + \hat{b}^\dagger_{k\lambda}(t)\mathscr{C}^*_{\mathbf{k}\lambda}(\mathbf{r})\right]. \quad (3)$$

The operators $\hat{b}^\dagger_{\mathbf{k}\lambda}$, $\hat{b}_{\mathbf{k}\lambda}$ of creation and annihilation, are Hermitian conjugated one to another.

In isotropic medium, the deformation potential determines the interaction energy operator of the electron system (exciton) with acoustic phonons

$$\hat{W}_d \equiv \hat{V}_{eph} =$$

$$= -\gamma\sum_{\mathbf{k},\lambda}\sqrt{\frac{N}{2m\hbar\omega_k}}\left(i\mathbf{e}_{\mathbf{k}}\cdot\mathbf{k}_{i\to f}\right)\hat{c}^\dagger_{f\sigma}\hat{c}_{i\sigma}\left(b_{\mathbf{k},\lambda}e^{i\mathbf{k}_\lambda\cdot\mathbf{r}} - b^+_{-\mathbf{k},\lambda}e^{-i\mathbf{k}_\lambda\cdot\mathbf{r}}\right). \quad (4)$$

Here, creation and annihilation operators $\hat{c}^\dagger_{f\sigma}, \hat{c}_{i\sigma}$ for electrons in excitons states $|\eta_{i\sigma}\rangle, |\eta_{f\sigma'}\rangle$ are fermionic operators: $\hat{c}^\dagger_{f\sigma}\hat{c}_{i\sigma} + \hat{c}_{i\sigma}\hat{c}^\dagger_{f\sigma'} = \delta_{if}\delta_{\sigma\sigma'}$. Now we mean the initial and final states as "exciton+phonon" states are described by vectors $|i\rangle = |n^{(i)}_{\mathbf{k},\lambda}\eta_{i\sigma}\rangle$, $|f\rangle = |n^{(f)}_{\mathbf{k},\lambda}\eta_{f\sigma'}\rangle$.

The operator $\hat{n}_{\mathbf{k}\lambda} = \hat{b}^\dagger_{\mathbf{k}\lambda}\hat{b}_{\mathbf{k}\lambda}$ corresponds to measurable physical quantity, which we denote by $n_{\mathbf{k}\lambda}$ and means a certain number of energy quanta in a given mode (phonons), the eigenstate is denoted as $|n_{\mathbf{k}\lambda}\rangle$. Then it will be fair to write

$$\hat{n}_{\mathbf{k}\lambda}|n_{\mathbf{k}\lambda}\rangle = \hat{b}^\dagger_{\mathbf{k}\lambda}\hat{b}_{\mathbf{k}\lambda}|n_{\mathbf{k}\lambda}\rangle = n_{\mathbf{k}\lambda}|n_{\mathbf{k}\lambda}\rangle.$$

They are characterized by the fact that the number of phonons in them is precisely defined, while the phase in them is absolutely undefined. This combination of operators, up to a factor, the dimension of energy, determines the operator of the energy of the crystal vibrations:

$$\hat{\mathscr{K}}^0 = \sum_{i=1}^N\left\{\frac{m}{2}\hat{\dot{\boldsymbol{\xi}}}_i^2 + \frac{\gamma}{2}\left(\hat{\boldsymbol{\xi}}_i - \hat{\boldsymbol{\xi}}_{i-1}\right)^2\right\} =$$

$$= \sum_{\mathbf{k}\lambda}\hbar\omega_k\left(\hat{b}^\dagger_{\mathbf{k}\lambda}\hat{b}_{\mathbf{k}\lambda} + \frac{1}{2}\right) = \sum_{\mathbf{k}\lambda}\hbar\omega_k\left(\hat{n}_{\mathbf{k}\lambda} + \frac{1}{2}\right). \quad (5)$$

Thus, there is a set of eigenvalues $\{n\}$ of the indicated operator and the corresponding set of states $\{|n\rangle_{\mathbf{k}\lambda}\}$ corresponding, as a solution of the Sturm–Liouville problem, to the condition of orthogonality $\langle m_{\mathbf{k}\lambda}|n_{\mathbf{k}\lambda}\rangle = \delta_{mn}$ and completeness $\sum_n |n\rangle_{\mathbf{k}\lambda}\langle n|_{\mathbf{k}\lambda} = \hat{1}$. We define here |ket⟩-vector of state $|n\rangle$ as an infinite-dimensional one-column matrix, and, accordingly, its conjugate ⟨bra|-vector $\langle n|$ as an infinite-dimensional one-row matrix, that is, the indicated property completeness is written as an infinite-dimensional unit matrix. The matrix elements of the operators of creation and annihilation of the energy quanta (phonon) in a given state are

$$\langle n_{\mathbf{k}\lambda} - 1|\hat{b}_{\mathbf{k}\lambda}|n_{\mathbf{k}\lambda}\rangle^\dagger = \langle n_{\mathbf{k}\lambda}|\hat{b}^\dagger_{\mathbf{k}\lambda}|n_{\mathbf{k}\lambda} - 1\rangle = \sqrt{n_{\mathbf{k}\lambda}}. \quad (6)$$

The considered basis of Fock states is convenient in those cases when the processes of interaction of a weakly intense deformation field with an electronic system (in exciton state), when we are dealing with one-phonon transitions inside material electrons, are described.

Next, we will discuss one-quantum transitions in crystals. Total Hamiltonian including "exciton-phonon" interaction is

$$\hat{\mathscr{K}} = \hat{\mathscr{K}}^0 + \hat{V} = \sum_{\mathbf{k}\lambda}\left\{\hbar\omega_k\left(\hat{b}^\dagger_{\mathbf{k}\lambda}\hat{b}_{\mathbf{k}\lambda} + \frac{1}{2}\right) + \right.$$

$$\left. + \left(-\frac{i}{\hbar}\right)(-1)e\gamma\sqrt{\frac{\hbar}{2m\omega_k N}}\,\mathbf{e}_{k\lambda}\cdot i\mathbf{k}\langle\eta_f|\hat{c}^\dagger_f\hat{c}_i|\eta_i\rangle\left\{\hat{b}^\dagger_{\mathbf{k}\lambda} + \hat{b}_{\mathbf{k}\lambda}\right\}\right\}. \quad (7)$$

Here Eq. (7) is obtained after averaging (4) on the electron components in the exciton polarization states, where the matrix elements $\langle \eta_f | \hat{V}_{eph} | \eta_i \rangle$ calculated over the initial and final states of the electronic system. Then we have a Hamiltonian of phonon subsystem with interaction, where instead of γ as in Eq. (4), we have the constant Ξ, calculated in Ref. [11], which, in addition to the deformation potential constant $\gamma = \partial^2 U(x)/\partial x^2$, (γ ~ 10 eV), also contains the contribution of the polarization potential, where P is the amplitude of the exciton polarization, $\mathfrak{E}$ is the Young's modulus, this field takes into account both the mechanical and exciton contributions:

$$\hat{\mathcal{H}} = \sum_{\mathbf{k}\lambda} \left\{ \hbar\omega_k \left(\hat{b}^{\dagger}_{\mathbf{k}\lambda} \hat{b}_{\mathbf{k}\lambda} + \frac{1}{2} \right) - \Lambda_{\mathbf{k}\lambda} \left\{ \hat{b}^{\dagger}_{\mathbf{k}\lambda}(t') + \hat{b}_{\mathbf{k}\lambda}(t') \right\} \right\},$$

$$\Lambda_{\mathbf{k}\lambda} = \left(-\frac{i}{\hbar} \right) ei\gamma \mathbf{e}_{k\lambda} \cdot \mathbf{k} \sqrt{\frac{\hbar}{2m\omega_k N}} \langle \eta_f | \hat{c}^{\dagger}_f \hat{c}_i | \eta_i \rangle =$$

$$= e\Xi \mathbf{e}_{k\lambda} \cdot \mathbf{k} \sqrt{\frac{1}{2m\hbar\omega_k N}},$$

$$\Xi = \gamma \left(1 + \frac{4\pi\gamma^2}{\omega \mathfrak{E}} |P|^2 \right). \tag{8}$$

To diagonalize Hamiltonian of phonon system in interaction (8), we seek a unitary transformation for the creation-annihilation operators in the following way

$$\hat{b}^{\dagger}_{\mathbf{k}\lambda} = B^{\dagger}_{\mathbf{k}\lambda} + u_{\mathbf{k}\lambda}{}^{*},$$
$$\hat{b}_{\mathbf{k}\lambda} = B_{\mathbf{k}\lambda} + u_{\mathbf{k}\lambda},$$
$$\left[B_{\mathbf{k}'\lambda'}, B^{\dagger}_{\mathbf{k}\lambda} \right] = \delta_{\mathbf{k}'\mathbf{k}} \delta_{\lambda'\lambda}. \tag{9}$$

Here function $u_{\mathbf{k}\lambda} = \Lambda_{\mathbf{k}\lambda} / (\hbar\omega_k)$ is real. Then, Hamiltonian (8) in new representation has a diagonal form

$$\hat{\mathcal{H}} = \sum_{\mathbf{k}\lambda} \hat{\mathcal{H}}_{\mathbf{k}\lambda},$$

$$\hat{\mathcal{H}}_{\mathbf{k}\lambda} = \hbar\omega_k \left(B^{\dagger}_{\mathbf{k}\lambda} B_{\mathbf{k}\lambda} + \frac{1}{2} \right) - \frac{2\left(\Lambda_{\mathbf{k}\lambda}\right)^2}{\hbar\omega_k}. \tag{10}$$

Such a transformation can be performed under each mode by using the following unitary transformation operator [18]

$$\hat{U}\left(\alpha_{\mathbf{k}\lambda}\right) = e^{\left(\alpha_{\mathbf{k}\lambda}{}^{*} \hat{b}^{\dagger}_{\mathbf{k}\lambda} - \alpha_{\mathbf{k}\lambda} \hat{b}_{\mathbf{k}\lambda}\right)},$$

$$\alpha_{\mathbf{k}\lambda} = u_{\mathbf{k}\lambda} = \frac{\Lambda_{\mathbf{k}\lambda}}{\hbar\omega_k}. \tag{11}$$

Appling this unitary transformation operator on to vacuum state of each mode, we get the new modes vacuum vector in the form:

$$\left| \nu_{\mathbf{k}\lambda} = 0 \right\rangle = \hat{U}\left(\alpha_{\mathbf{k}\lambda}\right) \left| n_{\mathbf{k}\lambda} = 0 \right\rangle = e^{\left(\alpha_{\mathbf{k}\lambda}{}^{*} \hat{b}^{\dagger}_{\mathbf{k}\lambda} - \alpha_{\mathbf{k}\lambda} \hat{b}_{\mathbf{k}\lambda}\right)} \left| n_{\mathbf{k}\lambda} = 0 \right\rangle =$$

$$= e^{-\frac{|\alpha_{\mathbf{k}\lambda}|^2}{2}} \sum_{\nu=0}^{\infty} \frac{\left(\alpha_{\mathbf{k}\lambda}\right)^{\nu}}{\sqrt{\nu!}} |\nu\rangle = |\alpha\rangle_{\mathbf{k}\lambda}. \tag{12}$$

It is easy to show, that this coherent state $|\alpha\rangle_{\mathbf{k}\lambda}$ has the following properties [18]:

1) It is the eigenvector of annihilation operator $\hat{b}_{\mathbf{k}\lambda} |\alpha\rangle_{\mathbf{k}\lambda} = \alpha_{\mathbf{k}\lambda} |\alpha\rangle_{\mathbf{k}\lambda}$.

2) An average number of phonons in this state is dependent on the interaction constant

$$\langle n \rangle_{\mathbf{k}\lambda} = \langle \alpha | \hat{b}^{\dagger} \hat{b} | \alpha \rangle_{\mathbf{k}\lambda} = |\alpha|_{\mathbf{k}\lambda}{}^2 = \left(\frac{\Lambda_{\mathbf{k}\lambda}}{\hbar\omega_k} \right)^2,$$

$$|\alpha\rangle_{\mathbf{k}\lambda} = e^{-\langle n \rangle_{\mathbf{k}\lambda}/2} \sum_{n=0}^{\infty} |n\rangle_{\mathbf{k}\lambda} \frac{\left(\sqrt{\langle n \rangle_{\mathbf{k}\lambda}} \right)^n}{\sqrt{n!}}. \tag{13}$$

3) A distribution function on the number of phonons in each mode for such coherent state is a Poisson distribution

$$W_{\mathbf{k}\lambda}(n) = e^{-\langle n \rangle_{\mathbf{k}\lambda}} \frac{\langle n \rangle_{\mathbf{k}\lambda}{}^n}{n!}. \tag{14}$$

4) These coherent states are non-orthogonal if correspond to different average numbers; nevertheless, they are fulfilled to the fullness property [18]:

$$\langle \alpha | \beta \rangle = \exp\left(-\left(\sqrt{\langle K \rangle} - \sqrt{\langle N \rangle} \right)^2 / 2 \right),$$

$$\left| \langle \alpha | \beta \rangle \right|^2 = \exp\left(-\left(\sqrt{\langle K \rangle} - \sqrt{\langle N \rangle} \right)^2 \right), \tag{15}$$

$$\frac{1}{\pi} \int |\alpha\rangle \langle \alpha | d^2\alpha = \sum_{n=0}^{\infty} |n\rangle \langle n| = \hat{1}. \tag{16}$$

The eigen energy of each mode for coherent state for Hamiltonian (10) is

$$\varepsilon_{\mathbf{k}\lambda} = \hbar\omega_k \left(\alpha_{\mathbf{k}\lambda} + \frac{1}{2} \right) - \frac{2\left(\Lambda_{\mathbf{k}\lambda}\right)^2}{\hbar\omega_k} = \Lambda_{\mathbf{k}\lambda} \left(1 - \frac{2\Lambda_{\mathbf{k}\lambda}}{\hbar\omega_k} \right) + \frac{\hbar\omega_k}{2},$$

where Eq.(11) for $\alpha_{\mathbf{k}\lambda}$ was taken into account. In contrary to the Fock-states, obtained coherent states are characterized by undetermined number of phonon in the given mode (only $\langle n \rangle_{\mathbf{k}\lambda}$ is known, see Eq. (13)), but exact determined phase. It is important to remind, that despite an operator of phase $\hat{\varphi}$ does not exist, two operators $\left(\hat{e}^{i\varphi} \right)$ and $\left(\hat{e}^{-i\varphi} \right)$ are used, their commutation relation is $\left[\hat{n}, \left(e^{i\varphi} \right) \right] = -\left(e^{-i\varphi} \right)$ [18,20].

3. TIME-DEPENDENT PROBLEM FOR PHONON COHERENT STATES

Let us consider a situation where the optomechanical interaction between optically pumped excitons and propagating acoustic waves explicitly depends on time, which is due to the polariton generation of excitons under the action of a laser pulse on the nanostructure, then instead Eq. (8) we have

$$\Lambda_{\mathbf{k}\lambda}(t) = e\Xi(t)\mathbf{e}_{k\lambda}\cdot\mathbf{k}\sqrt{\frac{1}{2m\hbar\omega_k N}}. \tag{17}$$

The general solution of the time-dependent Schrödinger equation

$$i\hbar\frac{\partial|\Psi_{\mathbf{k}\lambda}(t)\rangle}{\partial t} = \hat{\mathcal{H}}_{\mathbf{k}\lambda}(t)|\Psi_{\mathbf{k}\lambda}(t)\rangle$$

with the Hamiltonian

$$\hat{\mathcal{H}}_{\mathbf{k}\lambda}(t) = \hbar\omega_k\left(\hat{b}^{\dagger}_{\mathbf{k}\lambda}\hat{b}_{\mathbf{k}\lambda}+\frac{1}{2}\right)-\Lambda_{\mathbf{k}\lambda}(t)\left\{\hat{b}^{\dagger}_{\mathbf{k}\lambda}+\hat{b}_{\mathbf{k}\lambda}\right\}$$

has a form

$$|\Psi_{\mathbf{k}\lambda}(t)\rangle = C_{\mathbf{k}\lambda}(t)e^{\alpha_{\mathbf{k}\lambda}(t)b_{\mathbf{k}\lambda}{}^{\dagger}}e^{\beta_{\mathbf{k}\lambda}(t)b_{\mathbf{k}\lambda}}e^{\gamma_{\mathbf{k}\lambda}(t)b_{\mathbf{k}\lambda}{}^{\dagger}b_{\mathbf{k}\lambda}}|\Psi_{\mathbf{k}\lambda}(-\infty)\rangle, \tag{18}$$

where a vector $|\Psi_{\mathbf{k}\lambda}(-\infty)\rangle$ corresponds to the initial state of phonon system before acting the laser pulse onto the nanocrystal. The equation system for determination of coefficients $\alpha_{\mathbf{k}\lambda}(t)$, $\beta_{\mathbf{k}\lambda}(t)$, $\gamma_{\mathbf{k}\lambda}(t)$ and $C_{\mathbf{k}\lambda}(t)$ is obtained in general form by the same way as in Ref. [19]

$$\dot{\gamma}_{\mathbf{k}\lambda}(t) = -i\omega_{\mathbf{k}},$$

$$\dot{\beta}_{\mathbf{k}\lambda}(t) - i\omega_{\mathbf{k}\lambda}\beta_{\mathbf{k}\lambda}(t) = \frac{i\Lambda_{\mathbf{k}\lambda}(t)}{\hbar},$$

$$\dot{\alpha}_{\mathbf{k}\lambda}(t) + i\omega_{\mathbf{k}}\alpha_{\mathbf{k}\lambda}(t) = \frac{i\Lambda_{\mathbf{k}\lambda}(t)}{\hbar},$$

$$\frac{\dot{C}_{\mathbf{k}\lambda}(t)}{C_{\mathbf{k}\lambda}(t)} - \alpha_{\mathbf{k}\lambda}(t)\left(\dot{\beta}_{\mathbf{k}\lambda}(t) - \beta_{\mathbf{k}\lambda}(t)i\omega_{\mathbf{k}}\right) = -\frac{i}{2}\omega_{\mathbf{k}}. \tag{19}$$

The general solution of the time-dependent problem is presented in the quadrature's form

$$C_{\mathbf{k}\lambda}(t) = e^{-i\omega t/2}\times$$

$$\times\exp\left\{-\frac{1}{\hbar^2}\int_{-\infty}^{t}dt'\left(\Lambda_{\mathbf{k}\lambda}(t')e^{-i\omega t'}\int_{-\infty}^{t'}dt''\left(\Lambda_{\mathbf{k}\lambda}(t'')e^{i\omega t''}\right)\right)\right\},$$

$$\alpha_{\mathbf{k}\lambda}(t) = \frac{ie^{-i\omega_{\mathbf{k}}t}}{\hbar}\int_{-\infty}^{t}\Lambda_{\mathbf{k}\lambda}(t')e^{i\omega_{\mathbf{k}}t'}dt',$$

$$\beta_{\mathbf{k}\lambda}(t) = \frac{ie^{i\omega_{\mathbf{k}}t}}{\hbar}\int_{-\infty}^{t}\Lambda_{\mathbf{k}\lambda}(t')e^{-i\omega_{\mathbf{k}}t'}dt',$$

$$\gamma_{\mathbf{k}\lambda}(t) = -i\omega_{\mathbf{k}}t. \tag{20}$$

We consider the evolution of a phonon "wave packet" due to its appearance by interaction with an electronic wave packet generated by the exciton component of a polariton in the form $\Lambda_{\mathbf{k}\lambda}(t) = \Lambda_{\mathbf{k}\lambda 0}\exp(-|t|/\tau)$. We assume, that initial state of the phonon system corresponds to vacuum state and we use the initial conditions for the coefficients (20) in the following form

$$|\Psi_{\mathbf{k}\lambda}(-\infty)\rangle = |0\rangle_{\mathbf{k}\lambda},\ \alpha_{\mathbf{k}\lambda}(-\infty) = 0,$$

$$\beta_{\mathbf{k}\lambda}(-\infty) = 0,\ C_{\mathbf{k}\lambda}(-\infty) = 1. \tag{21}$$

With respect to initial conditions (21) we have the solution for the wave vector as follows

$$|\Psi_{\mathbf{k}\lambda}(t)\rangle = C_{\mathbf{k}\lambda}(t)e^{\alpha_{\mathbf{k}\lambda}(t)b^{\dagger}}e^{\beta_{\mathbf{k}\lambda}(t)b}e^{\gamma_{\mathbf{k}\lambda}(t)b^{\dagger}b}|0\rangle_{\mathbf{k}\lambda} =$$

$$= C_{\mathbf{k}\lambda}(t)\sum_{\nu=0}^{\infty}\frac{\alpha_{\mathbf{k}\lambda}{}^{\nu}(t)}{\nu!}b_{\mathbf{k}\lambda}{}^{\dagger\nu}|0\rangle = C_{\mathbf{k}\lambda}(t)\sum_{\nu=0}^{\infty}\frac{\alpha_{\mathbf{k}\lambda}{}^{\nu}(t)}{\sqrt{\nu!}}|\nu\rangle_{\mathbf{k}\lambda}, \tag{22}$$

obviously due to the equation

$$e^{\beta_{\mathbf{k}\lambda}(t)b}e^{\gamma_{\mathbf{k}\lambda}(t)b^{\dagger}b}|0\rangle_{\mathbf{k}\lambda} = e^{\beta_{\mathbf{k}\lambda}(t)b}\sum_{\nu=0}^{\infty}\frac{\gamma_{\mathbf{k}\lambda}{}^{\nu}(t)}{\nu!}\hat{n}_{\mathbf{k}\lambda}{}^{\nu}|0\rangle_{\mathbf{k}\lambda} =$$

$$= e^{\beta_{\mathbf{k}\lambda}(t)b}\cdot\hat{1}|0\rangle = |0\rangle_{\mathbf{k}\lambda}. \tag{23}$$

After integration with accounting of initial conditions (21), the coefficients have the form

$$\alpha_{\mathbf{k}\lambda}(t) = \frac{ie^{-i\omega_k t}}{\hbar}\Lambda_{\mathbf{k}\lambda 0}\left\{\frac{2\tau}{1+(\tau\omega_k)^2}-\frac{\tau e^{-\frac{t}{\tau}}e^{i\omega_k t}}{1-i\tau\omega_k}\right\}\underset{t\to\infty}{\to}$$

$$\underset{t\to\infty}{\to}\frac{ie^{-i\omega_k t}}{\hbar}\Lambda_{\mathbf{k}\lambda 0}\frac{2\tau}{1+(\tau\omega_k)^2},$$

$$\beta_{\mathbf{k}\lambda}(t) = \frac{ie^{i\omega_k t}}{\hbar}\Lambda_0\frac{2\tau}{1+(\omega_k\tau)^2},$$

$$\gamma_{\mathbf{k}\lambda}(t) = -i\omega t,$$

$$C_{\mathbf{k}\lambda}(t)\underset{t\to\infty}{\to}e^{-i\omega_k t/2}\exp\left\{-\frac{\Lambda_0^2}{2\hbar^2}\left[\frac{\frac{2}{\tau}}{\left(\frac{1}{\tau}\right)^2+\omega_k{}^2}\right]^2\right\}. \tag{24}$$

We prove that with accounting for Eq. (24) the normalization condition is fully satisfied:

$$\langle\Psi(t)||\Psi(t)\rangle = |C(t)|^2\sum_{\nu=0}^{\infty}\frac{|\alpha(t)|^{2\nu}}{\nu!} = |C(t)|^2 e^{|\alpha(t)|^2} = 1,$$

$$|C(t)|^2 = e^{-|\alpha(t)|^2},\Rightarrow|C(t)| = e^{-\frac{1}{2}|\alpha(t)|^2}. \tag{25}$$

Then, a heat flux of energy carried by phonons in coherent states for given mode **k**λ is

$$\langle j\rangle_{\mathbf{k}\lambda} = \hbar\omega_{\mathbf{k}} c_s \langle \Psi_{\mathbf{k}\lambda}(t)|\hat{n}_{\mathbf{k}\lambda}|\Psi_{\mathbf{k}\lambda}(t)\rangle =$$
$$= \hbar\omega c_s |\alpha_{\mathbf{k}\lambda}(t)|^2 |C_{\mathbf{k}\lambda}(t)|^2 e^{|\alpha_{\mathbf{k}\lambda}(t)|^2}, \quad (26)$$

where $c_s = \omega_k / k$ is a sound velocity (phase velocity of the given mode). With respect to the normalization condition (25), the spectral density of the energy flux is

$$\langle j\rangle_{\omega} = \frac{d\varepsilon}{d\omega} = \hbar\omega c_s \frac{g_s\omega^2}{(2\pi)^3 c_s^3}\frac{2\pi}{3}|\alpha_{\omega}(t)|^2 =$$
$$= \hbar\omega \frac{\omega^2}{(2\pi)^2 c_s^2}\left(\frac{2\Lambda_0\tau}{\hbar}\right)^2 \times$$
$$\times\left\{\frac{1}{\left(1+(\tau\omega)^2\right)^2} - \frac{e^{-t/\tau}\cos\left(\omega t + \operatorname{arctg}(\omega\tau)\right)}{\left(1+(\tau\omega)^2\right)^{3/2}} + \frac{e^{-2t/\tau}}{4\left(1+(\tau\omega)^2\right)}\right\}. \quad (27)$$

A total energy flux

$$J = \int_{-\infty}^{\infty}\langle j\rangle_{\omega} d\omega = \int_{-\varpi_D}^{\varpi_D} g_{\lambda}\frac{|\alpha(t)|^2}{4\pi^2 c_s^2}\hbar\omega\cdot\omega^2 d\omega = \left(\frac{\Lambda_0}{\tau\pi}\right)^2\frac{2}{\hbar c_s^2} g_{\lambda} \times$$
$$\times\left\{\left(1-\frac{e^{-2t/\tau}}{2}\right)\ln\left(1+(\tau\omega_D)^2\right) + \frac{1}{\left(1+(\tau\omega_D)^2\right)} - 1 + \frac{e^{-2t/\tau}}{2}(\tau\omega_D)^2\right\}. \quad (28)$$

With account of expression binding the sound velocity c_s and Debye frequency ω_D $c_s = \frac{\omega_D}{2\pi}\sqrt[3]{\left(\frac{4\pi}{3n}\right)}$ and ion concentration n in crystal, we have for the total energy flux

$$J = 3nc_s\frac{\Lambda_0}{(\tau\omega_D)^2}\frac{\Lambda_0}{\hbar\omega_D} \times$$
$$\times\left\{\left(1-\frac{e^{-2t/\tau}}{2}\right)\ln\left(1+(\tau\omega_D)^2\right) + \frac{1}{\left(1+(\tau\omega_D)^2\right)} - 1 + \frac{e^{-2t/\tau}}{2}(\tau\omega_D)^2\right\}. \quad (29)$$

The dependence of the total energy flux on the combination of the Debye frequency ω_D and the coherence time τ is presented in Figure 1. It is clearly seen that for certain ratios of these parameters, namely, $x = \tau\omega_D \sim 2$, the value of the total energy flux does not decay with time. Taking into account the Debye frequencies of acoustic phonons $\omega_D \sim 12.8$ THz and the characteristic coherence times $\tau \sim 10^{-13}$ s indicated in Ref. [21] for boron arsenide (BAs), it is clearly seen that they fit into the specified estimate, which can be associated with the regime of thermal superconductivity [14–17,21–24]. In Ref. [24], the dynamics of coherent optical phonons in tellurium after exposure to an intense femtosecond laser pulse is studied. The main mechanism of anomalous thermal phenomena in a material, up to a "non-temperature" phase transition, is considered to be the so-called DECP (Displacive Excitation of Coherent Phonons). It is shown that the time required for a carrier to diffuse over one absorption length (50 nm) is about 600 fs = 6×10^{-13} s, the characteristic frequencies at which resonant effects of thermal conductivity are observed are 3–3.6 THz. Our estimate of the parameter $x = \tau\omega_D$ for this system is x = 1.8–2.16, which corresponds to the undamped energy flow shown in Fig. 1. Moreover, in order not to resort to assessing the behavior of nanosized objects using reference values for the speed of sound in a crystal, or the Debye frequency obtained for macroscopic materials, you can use the microscopic value of the Debye frequency, as the maximum possible frequency in the propagation of harmonic excitation, corresponding to the minimum length wave equal to two a lattice constants of a particular nanocrystal: $\omega_D = \omega_{\max} = \frac{2\pi}{\lambda_{\min}}\sqrt{\frac{\gamma}{m}}a = \pi\sqrt{\frac{\gamma}{m}}$.

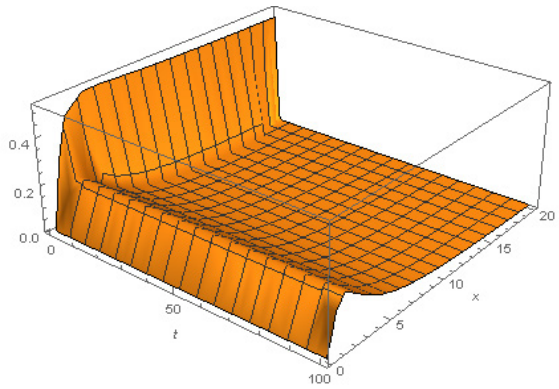

Fig. 1. Total energy flow as a function of dimensionless time $t = t/\tau$ (in units of decay time) and $x = \omega\tau$. At $x \sim 2$ the flow is maximum and the flow does not decay with time.

4. COHERENT HEAT FLUX AT FINITE TEMPERATURE

Let us now consider the problem for finite temperatures. Here it is necessary to take into account the entire continuum of modes across the entire spectrum of allowed frequencies. The spectral heat flux density, taking into account the finite temperature, must take into account the thermal distribution of the number of phonons in a given mode (Planck distribution function).

$$\langle j\rangle_{\omega T} = \left\langle\frac{d\varepsilon}{d\omega}\right\rangle_T = \hbar\omega c_s\frac{g_s\omega^2}{(2\pi)^3 c_s^3}\frac{2\pi}{3}|\alpha(t)|^2\left(\frac{1}{e^{\frac{\hbar\omega}{T}}-1}\right) =$$
$$= \hbar\omega\frac{\omega^2}{(2\pi)^2 c_s^2}\left(\frac{2\Lambda_0\tau}{\hbar}\right)^2\left\{\frac{1}{\left(1+(\tau\omega)^2\right)^2} + \frac{e^{-2t/\tau}}{4\left(1+(\tau\omega)^2\right)}\right\}\left(\frac{1}{e^{\frac{\hbar\omega}{T}}-1}\right) =$$

$$=\left(\frac{\hbar}{\tau T}\right)(\omega\tau)T\frac{(\tau\omega)^2}{(2\pi)^2 c_s^2}\left(\frac{2\Lambda_0}{\hbar}\right)^2\left\{\frac{1}{\left(1+(\tau\omega)^2\right)^2}+\frac{e^{-2t/\tau}}{4\left(1+(\tau\omega)^2\right)}\right\}\left(\frac{1}{e^{\left(\frac{\hbar}{\tau T}\right)(\tau\omega)}-1}\right), \quad (30)$$

The spectral flux density accounts either the Planck equilibrium distribution and orders of magnitude for the coherence time τ determined by the characteristic relaxation times of the electronic system in a solid, that is, $\tau\sim10^{-13}$ s. A parameter included in the Planck distribution function $\hbar/T$ in the temperature range from 300 to 1000 K, in order of magnitude corresponds to the range of values 10^{-14}–10^{-15} s. The spectral flux density taking into account the Planck equilibrium distribution is presented in Figure 2, where denotations $t=t/\tau$, $x=\omega\tau$ are used. For a given phonon relaxation time, there is a resonance frequency $\omega^*\sim3/2\tau$, when the spectral flux density does not decay with time, which can be associated with the phenomenon of thermal superconductivity observed in experiments [24].

Let us consider the total flow taking into account the temperature in the low-temperature limit, $T\le\Lambda_0$ (here the temperature is written in the energy scale). In the subsequent integral the momentum relaxation time is quite long $\tau>>\hbar/T$ due to $\tau\sim10^{-13}$, $\hbar/T\sim10^{-14}-10^{-15}$, $\hbar/(\tau T)\sim0.01-0.1$. In other words, we are dealing with a quasi-adiabatic interaction, when the system can be described by the Planck equilibrium distribution function. The condition of a small gradient means that the inequality $\tau c_s<<l$ is satisfied, where l is the spatial scale of temperature change. In other words, the temperature change itself occurs over relatively long times, thus, the integration of the spectral flux density over frequency, taking into account temperature, can be performed as at a constant temperature.

$$J=\int_0^{\varpi_D}\langle j\rangle_\omega\left(\frac{1}{e^{\frac{\hbar\omega}{T}}-1}\right)d\omega=\int_0^{\varpi_D}\frac{|\alpha(t)|^2}{4\pi^2c_s^2}\left(\frac{\hbar\omega}{e^{\frac{\hbar\omega}{T}}-1}\right)\omega^2d\omega=$$

$$=\hbar\left(\frac{\Lambda_0\tau}{\hbar\pi c_s}\right)^2\int_0^{\varpi_D}\left\{\frac{1}{\left(1+(\tau\omega)^2\right)^2}+\frac{e^{-2t/\tau}}{4\left(1+(\tau\omega)^2\right)}\right\}\left(\frac{1}{e^{\frac{\hbar\omega}{T}}-1}\right)\omega^3d\omega. \quad (31)$$

1. Low temperature limit: $\tau\omega_D\prec2$, $\hbar\omega_D/T>>1$.

$$J_{LT}\approx\int_0^{\varpi_D}\langle j\rangle_\omega\left(\frac{1}{e^{\frac{\hbar\omega}{T}}-1}\right)d\omega=$$

$$=\left(\frac{\tau\Lambda_0}{\pi\hbar}\right)^2\frac{\hbar}{\tau^4c_s^2}\int_0^{\infty}\left(\frac{1}{e^{\frac{\hbar\omega\tau}{\tau T}}-1}\right)\frac{(\tau\omega)^3}{\left(1+(\tau\omega)^2\right)^2}d(\tau\omega)\approx$$

$$\approx\frac{2}{5}nc_s\frac{(\pi T)^4}{(\hbar\omega_D)^3}\left(\frac{\tau\Lambda_0}{\hbar}\right)^2, \quad (32)$$

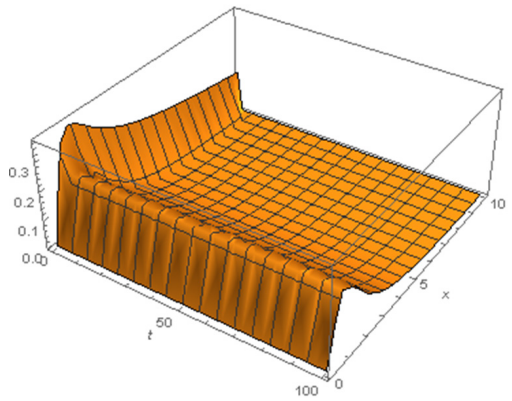

Fig. 2. The spectral flux density for the finite temperature. Here denotations $t=t/\tau$, $x=\omega\tau$ are used. For a given phonon relaxation time, there is a resonance frequency $\omega^*\sim3/2\tau$, when the spectral flux density does not decay with time.

The result is the same as for thermal phonons in the absence of coherence, if the coherence factor is $\tau\Lambda_0/\hbar\to1$.

2. High temperature limit: $\tau\omega_D\prec2$, $\hbar\omega_D/T\sim1$.

$$J_{HT}=\int_0^{\infty}\frac{\langle j\rangle_\omega}{e^{\frac{\hbar\omega}{T}}-1}d\omega=$$

$$=\frac{3}{2}\frac{nTc_s}{\tau\omega_D}\left(\frac{\Lambda_0}{\hbar\omega_D}\right)^2\left\{\pi+\frac{e^{-2t/\tau}}{2}\left[(\tau\omega_D)^2-\ln\left(1+(\tau\omega_D)^2\right)\right]\right\}. \quad (33)$$

We arrive at the classical linear dependence on temperature, which occurs in the high-temperature limit in a macroscopic crystal. However, here there is also a coherent contribution directly related to the coherence time τ, decayed exponentially with time, which is quite consistent with the conclusions of Refs. [9,23,24] for the high temperature limit taking into account the coherence of phonons.

5. RESULTS AND DISCUSSIONS

Pulsed excitation and phase-sensitive detection of coherent phonons and phonon-polaritons provide detailed insight into the dynamic properties of matter. Experiments [24] based on optical pumping methods with femtosecond time resolution make it possible to simultaneously determine the amplitude and phase of coherent grating motion. Frequencies in the terahertz range and dephasing times in the picosecond range are obtained with high accuracy, especially in semiconductors and semiconductor heterostructures, where the coherent phonon mode and free carriers are excited simultaneously, which carries important information about the interaction of carriers with phonons far from equilibrium. Experiments on the generation of LO phonons are carried out in Sb, where the A mode is similarly manipulated. In mixed BiSb crystals, Bi-Bi, Bi-Sb and Sb-Sb vibrations can be amplified and suppressed using a series of femtosecond

pulses. In all of these experiments, the general pattern discovered in our work is observed, namely: the very effect of amplification of LO phonons with undamped thermal conductivity occurs in various materials when the criterion $x = \tau\omega^* \sim 2$ is met, where τ is the temporal coherence, ω^* is the frequency at which the maximum spectral energy density occurs.

In this work $\tau = 0.25$ ps $= 0.25\times10^{-12}$ s, $\omega^* = 8.8$ THz $= 8.8\times10^{12}$ Hz, $\tau\omega^* = 2.2$. For bulk GaAs: $\tau = 0.25$ ps, $\omega^* = 9$ THz, $x = \tau\omega^* = 2.25$, for GaAs/Al0.3Ga0.7As superlattice: $\tau = 0.2$ ps, $\omega^* = 8.5$ THz, $x = \tau\omega^* = 1.7$. For isotropic Te crystal: $\omega^* = 3.5$ THz, $\tau = 0.6$ ps, $x = \tau\omega^* = 2.1$. For anisotropic Te crystal: $\omega^* = 4.2$ THz, $\tau = 0.5$ ps, $x = \tau\omega^* = 2.1$.

In the terahertz emission experiment, for InP crystal: wide peak at $\omega^* = 1$ THz, $\tau = 2$ ps, $x = \tau\omega^* = 2$.

In a study of coherent phonon dynamics in a Te single crystal excited by amplified CPM laser pulses with a photon energy of 2 eV:
for Te crystal: at $\omega^* = 3.6$ THz, $\tau = 0.75$ ps, $x = \tau\omega^* = 2.7$; for HTSC material, $YBa_2Cu_3O_7$-thin film: Ba mode in the CuO_2 plane: $\omega^* = 3.6$ THz, $\tau = 0.7$ ps, $x = \tau\omega^* = 2.52$; Cu mode (2) in the CuO_2 plane: $\omega^* = 4.2$ THz, $\tau = 0.7$ ps, $x = \tau\omega^* = 2$.

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УДК 538.94:538.931:538.975:538.913:538.911

О существовании фононных когерентных состояний в наноматериалах

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Аннотация. Работа посвящена описанию переноса энергии тепловыми возбуждениями с учетом их когерентности в наноразмерных системах при теплоотводе. Предложен общий формализм теплопроводности, учитывающий, как обычную фононную модель теплопередачи, так и образование когерентных шредингеровских состояний колебательной системы. Аналитически получено точное решение нестационарной задачи с произвольными начальными условиями. Показано, что при определенных соотношениях констант, характеризующих взаимодействие фононов с электронной подсистемой, тепловой поток в кристалле не затухает со временем.

Ключевые слова: когерентные состояния; когерентные волны; когерентный тепловой поток; нанокристаллы; тепловая сверхпроводимость