1. INTRODUCTION

The problem of heat transfer through a boundary of two substances currently attracts attention of researchers in view of its great importance for technical applications [1]. Kapitza [2] was the first who discovered that the temperature undergoes a jump at the boundary of two substances in the presence of the heat flux. The proportionality coefficient between the value of the heat flux and temperature jump is called the boundary thermal resistance (the Kapitza resistance). Initially, the temperature jump was found at the interface between liquid helium and a solid, and this case was investigated (see [3] and references therein). However, important experimental results on the thermal resistance at the interface between two crystals were also obtained in recent years [4–7]. In numerous studies, which are devoted to the theory of boundary thermal resistance [8–11], the heat flux through the boundary at given temperatures of substances is calculated; in this case, the distribution function of heat carrier particles in the substance is assumed to be the equilibrium distribution function with the corresponding temperature. However, the presence of a constant heat flux through the interface means the presence of the same flux in the substance; therefore, the distribution function differs from the equilibrium one. In this article, we introduce distribution functions and boundary conditions for distribution functions as was done for the homogeneous substance in the presence of the temperature gradient, i.e., by the Enskog–Chapman method [12]. It turns out that this leads to substantial corrections to the calculated boundary thermal resistance for the interface between two crystals. The authors of [13] consider oscillations in the chain consisting of two semi-infinite chains with one additional bond between them at the interface. If a plane wave falls onto the interface from one side, the transmitted and reflected waves appear. The problem of finding the amplitudes of the transmitted and reflected waves allowing for conditions at the interface between two media is completely solved in [13]. Further, the heat flux is found from the following considerations, which in essence coincide with those used in the acoustic mismatch model (AMM). Let us assume that the chain oscillates at a specific frequency. The number of phonons of this frequency is uniquely related to the temperature by the Bose–Einstein distribution function so that we can formally speak about the temperature in a system, the state of which is completely determined by the occupation number for one state only. We follow this approach in our analysis. We consider the model which assumes two solutions at a specific frequency. They correspond to a wave striking the interface from the left and from the right. Taking into account that the temperature calculated from the occupation numbers of phonons incident from the left (from the right) is the temperature of the substance on the left (on the right), we determine the heat flux through the interface as the sum of energy fluxes associated with each of phonons of the system, linearize over the temperature difference, and then integrate over all frequencies. As a result, we obtain the analog of the Landauer formula. However, in reality we cannot identify temperatures of such states with temperatures of substances.

2. INCORRECTNESS OF THE AMM FORMULA FOR A LARGE TRANSMISSION COEFFICIENT

Let us consider a simplest model of the interface between two crystals already described in the Introduction—a single one-dimensional chain, which is
characterized by the set of elastic constants $\beta_1$, $\beta_2$, and $\beta$ determining the interactions between the atoms inside the media and at the interface. Atomic weights of substances are denoted as $m_1$ and $m_2$ (see figure), and interatomic distances are denoted as $a_1$ and $a_2$. We neglect the anharmonicity of vibrations. If a plane wave of specified frequency $\omega$ and unit amplitude falls on the interface, then the reflected wave with amplitude $A$ and transmitted wave with amplitude $B$ appear. Vibrations of this type will be further referred to as coupled vibrations. The following properties of the solution are substantial for the further consideration: $|A_1| = |A_2| = |A|$, $|B_1|^2 = (v_1/v_2)(1 - A^2)$, and $|B_2|^2 = (v_2/v_1)(1 - |A|^2)$, where lower indices 1 and 2 mean that the wave falls onto the interface from the left or from the right, respectively; and $v_j$ are the group velocities. Presented relationships express the conservation of a heat flux. It is important that $A^2 + B^2 \neq 1$, i.e., squares of amplitudes of the reflected and transmitted waves cannot be interpreted as probabilities of phonon reflection and transmission. In [13], the heat flux is found by the formula

$$q = \Delta T \frac{1}{2\pi} \int_0^\infty \frac{\partial n_\omega}{\partial T} \hbar \omega d\omega,$$  \hspace{1cm} (1)

where $t = 1 - |A|^2$ is the transmission coefficient, $T \approx T_1 \approx T_2$ is the substance temperature, and $\Delta T = T_1 - T_2 \ll T$ is the temperature difference. It should be noted that although the integration is formally performed from zero to infinity, the transmission coefficient in reality turns to zero for frequencies exceeding the maximally possible vibration frequency in one of the substances: $\omega > \min[\omega_{m1}, \omega_{m2}]$. The energy transfer by phonons with frequencies $\max[\omega_{m1}, \omega_{m2}] > \omega > \min[\omega_{m1}, \omega_{m2}]$ is possible only allowing for inelastic processes [14]. One can readily verify that the calculation by formula (1) is incorrect at $t \approx 1$. Indeed, let us apply formula (1) for the uniform chain, i.e., for the case $\beta_1 = \beta_2 = \beta$, $m_1 = m_2$, and $a_1 = a_2$. It is evident that $t = 1$ in this case and no temperature jump will be at the “interface” since temperature jumps are impossible in the homogeneous medium. However, formula (1) gives the finite heat flux for the temperature jump, which was for the first time noted as far back as in [9]. We can expect that a multiplier of the form $1/(1 - t)^\alpha$ will appear in the integrand in the correct formula; in this case, the finite temperature jump in the homogeneous medium leads to an infinite heat flux or, simpler speaking, the temperature jump is impossible. For a simpler model of two coupled harmonic strings, such a formula was for the first time derived by van den Brink [15], and the value of $\alpha$ proved to be equal to unity. This author also proposed the method of derivation based on the modified Green’s function, and the result turned out to be exactly the same (for modern applications of the Green’s function method for calculating the Kapitza conductance, see [16] and references therein). Further, we derive the formula for the calculation of the Kapitza thermal resistance using the modified Enskog–Chapman method. In order to understand the error in the derivation of formula (1), let us come back to the consideration of the uniform chain. Two solutions occur for the specified frequency: the wave traveling from the left to the right and the wave traveling from the right to the left. The state of the system at the specified frequency is determined by two occupation numbers: $n_\rightarrow$ and $n_\leftarrow$, respectively. Following the scheme presented in Introduction, temperatures from the left and from the right should be determined by equations

$$n_\rightarrow = \frac{1}{\exp(\frac{\hbar \omega}{T_1}) - 1}, \quad n_\leftarrow = \frac{1}{\exp(\frac{\hbar \omega}{T_2}) - 1}.$$  \hspace{1cm} (2)

In this case, in the presence of the heat flux $n_\rightarrow \neq n_\leftarrow$; correspondingly, $T_1 \neq T_2$; i.e., the temperature jump occurs in the homogeneous medium, which is physically incorrect as it was already noted. In this situation, following Enskog and Chapman, the temperature should be determined using equation

$$n_0 = \frac{n_\rightarrow + n_\leftarrow}{2} = \frac{1}{\exp(\frac{\hbar \omega}{T}) - 1},$$  \hspace{1cm} (3)

while the heat flux—using equation

$$q = \hbar \omega \nu(\chi_\rightarrow - \chi_\leftarrow),$$  \hspace{1cm} (4)

where $\nu$ is the group velocity, $\chi_\rightarrow = n_\rightarrow - n_0$, and $\chi_\leftarrow = n_\leftarrow - n_0$ is the nonequilibrium additive to the distribution function, which satisfies condition $\chi_\rightarrow + \chi_\leftarrow = 0$. In this case, the temperature along the entire chain is constant even in the presence of the heat flux, as it should be, neglecting vibration anharmonicity. Indeed, in this case, there is nothing for phonons to scatter off, and the thermal conductance is infinite.
3. COUPLING OF THE DISTRIBUTION FUNCTIONS AT THE INTERFACE

Let us return to the discussion of the chain with the interface and will again consider vibrations at a specific frequency. It was already noted that two solutions in the form of coupled modes occur in this case; however, we cannot calculate the temperature of substances starting from the occupation numbers of these modes: this would be equivalent to the calculation of the temperature by the occupation number of phonons propagating only towards the interface. We could introduce a separate temperature for vibrations near the surface, i.e., coupled modes, as was done in [10]. With such an approach, their temperature differs from temperatures of each of substances, and heat transfer occurs through inelastic collisions of coupled modes with usual phonons. This method is, however, far from being physically transparent and is extremely unwieldy. An attempt to analyze the equations within this approach requires rough approximations. Instead, we will attempt to extend the determination of the temperature within the Enskog–Chapman approach to the case under consideration, i.e., to calculate the temperature through the arithmetic mean of occupation numbers of phonons propagating to the interface and from the interface both from the left and from the right. To do this, let us come from the basis of coupled modes to the basis of ordinary plane waves. Let us introduce \( N_1 \) and \( N_2 \)—occupation numbers of coupled modes, \( n_1 \) and \( n_{1e} \)—occupation numbers of phonons falling onto the interface and moving from the boundary in the substance from the left, and \( n_2 \) and \( n_{2e} \)—the same for phonons in the substance to the right from the interface. We should pass from \( N_1, N_2 \) to \( n_{1e}, n_{1e}, n_{2e}, n_{2e} \). For this purpose, let us express the vibrational amplitudes of the atoms to the left and to the right from the interface through the birth and annihilation operators by two different methods. Let us denote \( \hat{C}_1 \) and \( \hat{C}_2 \)—annihilation operators of coupled modes; \( \hat{c}_{1e} \) and \( \hat{c}_{1e} \)—annihilation operators of phonons propagating to the right and to the left, respectively, in the substance on the left; \( v_1 \) and \( v_2 \)—normalization constants for coupled modes; and \( \eta_1 \) and \( \eta_2 \)—for ordinary phonons to the left and to the right from the interface, respectively. The displacement operator of the \( n \)th atom to the left from the boundary is

\[
\hat{u}_n = v_1 \hat{C}_1 (e^{-ik_{1e}n} + A e^{ik_{1e}n}) + v_2 \hat{C}_2 B_2 e^{ik_{1e}n}
\]

and

\[
\hat{u}_n = v_1 \hat{c}_{1e} (e^{-ik_{1e}n}) + (v_1 \hat{A} \hat{C}_1 + v_2 \hat{B}_2 \hat{C}_2) e^{ik_{1e}n},
\]

on the other hand,

\[
\hat{u}_n = \eta_1 \hat{c}_{1e} (e^{-ik_{1e}n}) + \hat{c}_{1e} e^{ik_{1e}n}.
\]

Comparing (5) and (6), we find that

\[
\hat{c}_{1e} = (v_1/\eta_1) \hat{C}_1, \\
\hat{c}_{1e} = (v_1/\eta_1) A \hat{C}_1 + (v_2/\eta_2) B_2 \hat{C}_2.
\]

Now, we have

\[
n_{1e} = \langle n_{1e} \rangle_{n_{1e} n_{2e} n_{2e}} \langle n_{1e} n_{2e} n_{2e} \rangle = (v_1/\eta_1)^2 \langle n_1 N_2 \rangle \langle C_1 C_1 N_1 N_2 \rangle = (v_1/\eta_1)^2 N_1.
\]

Having done similar calculations for other occupation numbers, we derive

\[
\begin{align*}
n_{1e} &= (v_1/\eta_1)^2 N_1 \\
n_1 &= (v_1/\eta_1)^2 |A|^2 N_1 + (v_2/\eta_1)^2 |B_2|^2 N_2 \\
n_{2e} &= (v_2/\eta_2)^2 N_2 \\
n_2 &= (v_2/\eta_2)^2 |A|^2 N_2 + (v_1/\eta_2)^2 |B_1|^2 N_1.
\end{align*}
\]

Finally, we derive

\[
\begin{align*}
n_{1e} &= |A|^2 n_{1e} + (\eta_1/\eta_2)^2 |B_2|^2 n_{2e} \\
n_{2e} &= (\eta_1/\eta_2)^2 |B_1|^2 n_{1e} + |A|^2 n_{2e}.
\end{align*}
\]

Thus, we derived coupling conditions of distribution functions at the interface. Appropriate conditions, which associate normalization constants \( \eta_1 \) and \( \eta_2 \), will be derived below. We can now introduce

\[
n_{i0} = n_{1e} + n_{2e},
\]

on the other hand,

\[
n_{i0} = \frac{1}{\exp \frac{\hbar \omega}{T} - 1},
\]

where index \( i \) denotes substance 1 (on the left) and 2 (on the right). This is precisely the desired definition of temperatures for the chain with the interface. From the mathematical viewpoint, the system of equations (10) means the following. The space of states of the chains, which vibrates at a fixed frequency, is two-dimensional. When we pass into the basis of four ground states, the states, which can be implemented in reality, form a plane in the four-dimensional space. The system of equations (10) represents the equations of this plane. From the physical viewpoint, the system of equations (10) expresses a simple consideration that phonons moving from the interface are summed from those that fell onto the interface from the same side and reflected, and those that fell onto the interface from another side and transmitted through it. The fact that just squares of amplitudes stand in coefficients is also clear beforehand since the number of phonons in this mode is proportional to the vibrational energy of this mode, while the energy is proportional to the
square of the amplitude. Thus, it is seen that the procedure of broadening the basis, which can initially seem to be somewhat artificial, has the quite simple and illustrative physical sense. It also enables to spread the definition of temperatures for nonequilibrium systems accepted in kinetics to lattice vibrations near the interface between two crystals.

4. KAPITZA RESISTANCE

Equations (10)–(12) form the closed system of equations. The solution is performed in the reverse order. Knowing the temperatures, we find \( n_{i\omega} \); substituting (12) into (11), we find \( n_{1\omega} \) and \( n_{2\omega} \), substitute into (10), and determine \( n_{1\varepsilon} \) and \( n_{2\varepsilon} \). Knowing all occupation numbers, we find the heat flux. Introducing notation \( \chi_i = \chi_{\rightarrow} - \chi_{\leftarrow} = n_{i\rightarrow} - n_{i\leftarrow} \), we derive

\[
\chi_1 = \frac{1}{A_2} \left[ (1 - A^2)n_{10} - (\eta_2/\eta_1)^2 B_1^2 n_{20} \right],
\]

\[
\chi_2 = \frac{1}{A_2} \left[ (\eta_1/\eta_2)^2 B_2^2 n_{10} - (1 - A^2)n_{20} \right].
\]

(13)

The heat flux should be identical on both sides of the interface. From here,

\[
v_1 \chi_1 = v_2 \chi_2
\]

(14)

or

\[
v_1 [(1 - A^2)n_{10} - (\eta_2/\eta_1)^2 B_1^2 n_{20}] = v_2 [(\eta_1/\eta_2)^2 B_2^2 n_{10} - (1 - A^2)n_{20}].
\]

(15)

Equation (15) is fulfilled for any values of \( n_{10} \) and \( n_{20} \), from here it follows that

\[
(\eta_1/\eta_2)^2 B_1^2 = (v_1/v_2)t, \quad (\eta_2/\eta_1)^2 B_2^2 = (v_2/v_1)t.
\]

(16)

Using \( B_2^2 = \frac{v_1}{v_2} t \) and \( B_1^2 = \frac{v_2}{v_1} t \), we find

\[
\eta_1 = \eta_2.
\]

(17)

Substituting (17) into (13), we derive

\[
\chi_1 = \frac{1}{v_1^2} \frac{t}{1 - t} \left[ v_1 n_{10} - v_2 n_{20} \right],
\]

\[
\chi_2 = \frac{1}{v_2^2} \frac{t}{1 - t} \left[ v_1 n_{10} - v_2 n_{20} \right].
\]

(18)

Here, it is convenient to pass to the continuous limit substituting \( n_{i\omega} \rightarrow n_{i\omega} dk/2\pi \), and then, using \( v = \frac{do}{dk} \),

\[
\frac{do}{dk} n_{i\omega} dk = n_{i\omega} do.
\]

After this, we can linearize the expression with respect to the temperature difference under the assumption of its smallness. In order to find the total heat flux, we should substitute (18) into (4) and integrate over all frequencies. Finally, we derive

\[
q = \Delta T \frac{1}{2\pi} \int_0^\infty \frac{\hat{\nu}_n^0}{1 - t \partial T} \hat{\nu}_\omega d\omega.
\]

(19)

It is seen that the obtained formula at small \( t \) transforms into the formula from [13] similar to AMM. A similar formula was derived in [15] starting from other considerations. The presented derivation method using coupling conditions of distribution functions at the interface is convenient by the fact that it is generalized to a three-dimensional case by the introduction of additional indices. Expression (19) is derived under a strong simplifying assumption that Eq. (11) is fulfilled for each frequency separately. In reality, a much weaker integral relation

\[
\int n_{i\omega} \hat{\nu}_\omega d\omega = \int \frac{n_{i\rightarrow} + n_{i\leftarrow}}{2} \hat{\nu}_\omega d\omega
\]

(20)

takes place.

Solving the problem under this condition, we derive the following expression for the heat flux:

\[
q = \Delta T \frac{1}{2\pi} \int_0^\infty \frac{\hat{\nu}_n^0}{1 - t \partial T} (1 + f(\omega)) \hat{\nu}_\omega d\omega.
\]

(21)

It differs from (19) by integrand multiplier \( (1 + f(\omega)) \). In this case, \( f(\omega) \) is the alternating function of the order of unity. Multiplier \( \frac{1}{1 - t} \) is retained. Finding \( f(\omega) \) requires the joint solution of the Boltzmann equation for phonons on both sides of the interface with the coupling conditions of distribution functions (10), (17)

\[
\left\{
\begin{array}{l}
n_{1\rightarrow} = |A|^2 n_{1\rightarrow} + |B_1|^2 n_{2\varepsilon-}, \\
n_{2\leftarrow} = |B|^2 n_{1\rightarrow} + |A|^2 n_{2\varepsilon-}.
\end{array}
\right.
\]

(22)

This is the generalization of the Enskog–Chapman method for the case of two crystals. Expression (19) can be considered as the approximate evaluation. Thus, the use of the modified Enskog–Chapman method enables the derivation of the more exact expression for the Kapitza thermal resistance.

5. COMPARISON WITH THE EXPERIMENT

Let us try to perform the approximate comparison with the experiment. The authors of [4] compare the results of the experiment and calculation using the AMM, in which formula (1) is used, which is generalized to the three-dimensional case (integration is performed over the components of the wave vector of phonons, which is parallel to the boundary; transmission coefficient \( t \) is determined in a more complex
Kapitza thermal conductance for interfaces of different metals with diamond at room temperature (in units of 10^3 W cm^{-2} K^{-1})

| Metal | 1/(1 – t) | Experiment | AMM | AMM (corr.) |
|-------|-----------|------------|-----|------------|
| Pb    | 2.5       | 3          | 0.05| 0.125      |
| Au    | 50        | 4          | 0.4 | 20         |
| Al    | 2.5       | 5          | 5   | 12.5       |
| Ti    | 10        | 9          | 7   | 70         |

In the last column, the results of calculation with the use of the AMM are multiplied by 1/(1 – t).

Since (19) differs from (1) only by multiplier \( \frac{1}{1 – t} \), if \( t \) is independent of \( \omega \), we can simply addi-
ionally multiply the result of calculation using the AMM to the corresponding multiplier. The transmission coefficient at low frequencies can be found from the elasticity theory [4]

\[
t(0) = \frac{4Z_1Z_2}{(Z_1 + Z_2)^2}.
\]

Here, \( Z_1 \) is the so-called acoustic impedance; \( Z_i = v_i\rho_i \), where \( v_i \) is the sound velocity and \( \rho_i \) is the density in the \( i \)th substance. Since the transmission coefficient in reality decreases as the frequency increases (which is taken into account in [4]), \( \frac{1}{1 – t} \) also decreases so that the result (see table) turns out overestimated.

The data presented in the table for gold, aluminum, and titanium, as expected, turned out to be overesti-
mated. This suggests that, for interfaces between materials possessing substantially different acoustic impedances, one should take into account the inelastic energy transfer from phonons to electrons, for instance, with the inclusion of the mechanisms proposed in [11, 14].

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