

# Transport phenomena at a gas-solid interface due to propagation of surface sound

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Transport processes are considered in a single nonequilibrium gas-solid system in the case of propagation of a high-frequency ( $\omega_0\tau_p \gg 1$ , where  $\omega_0$  is the frequency of sound and  $\tau_p$  is the thermal phonon lifetime) surface acoustic wave in a solid. Absorption (creation) of single acoustic phonons by gas molecules when these molecules are scattered by the surface is the microscopic mechanism of interphase nonlocal transport effects (transport in one medium dependent on a nonequilibrium state of another medium) which occur under these conditions. A kinetic theory of a single nonequilibrium gas-solid system is developed and it is shown that, in particular, the dependence of the resultant energy flux in a gas on the mass of molecules and temperature is fundamentally different from the corresponding dependence obtained within the framework of the traditional mechanism of the interaction of gas molecules with a surface which is in a local equilibrium but vibrates under the action of the propagating sound. The theory has made it possible to explain for the first time the experimental data on the influence of a gas on the absorption of surface sound. A study has also been made of a new macroscopic motion of the gas along the surface, which is due to the drag of molecules by acoustic phonons. In the case of propagation of surface sound along a wall of a channel the velocity of flow of collisionless gas is proportional to  $\omega_0^{1/2}$ , is independent of the mass of the molecules, and under typical experimental conditions can reach an easily measurable value of the order of tens of centimeters per second.

## 1. INTRODUCTION

It is shown in Ref. 1 that under nonisothermal conditions the transport of heat and mass at a gas-solid interface may be strongly nonlocal: the processes of transport in one medium may depend on a nonequilibrium state of the other medium. We shall develop a theory of interphase nonlocal transport phenomena in a gas-solid system which appear as a result of a different type of nonequilibrium representing propagation of a surface acoustic wave in a solid. The absorption of sound by a gas represents an interphase transport effect if, as assumed below, it is associated with the absorption of acoustic phonons by gas molecules when these molecules are scattered by the surface. This approach has made it possible to explain for the first time the available experimental data on the influence of a gas on the absorption of surface sound and to predict macroscopic motion of a gas along a surface due to the drag of molecules by acoustic phonons.

When sound propagates in a solid, its wavelength is considerably greater than the interatomic distances or than the radius of action of molecule-surface forces right up to frequencies  $\omega_0 \sim 10^{11} - 10^{12}$  Hz. It would therefore seem that even at high frequencies a natural approach to the interaction of molecules of sound would be to consider that interaction as the scattering by a vibrating macroscopic element of the surface. However, the experimental data<sup>2</sup> on the dependence of  $\gamma_2 = \gamma - \gamma_1$  ( $\gamma$  is the absorption coefficient of surface acoustic waves and  $\gamma_1$  is the value of  $\gamma$  in the limit  $p \rightarrow 0$ ) on the frequency of sound  $\omega_0$  and on the mass of gas molecules  $m$  can be described (see Sec. 4 below) by a kinetic theory of a gas bounded by a surface vibrating macroscopically due to propagation of sound only at frequencies in the range  $(\omega_0/2\pi) < 10^9$  Hz. The experimental results ob-

tained at higher frequencies cannot be explained by this theory, which postulates a local equilibrium of a solid. On the other hand, at frequencies  $(\omega_0/2\pi) \gtrsim 10^9$  Hz and at room temperature the frequency of collisions of thermal phonons with one another ( $\tau_p^{-1}$ ), typical of problems on the absorption of sound in solids, is comparable with the frequency of sound.<sup>3</sup> Therefore, the mechanism of the scattering of molecules by a macroscopically vibrating local-equilibrium element of the surface may become inapplicable: the scattering law may generally depend not only on temperature, but also on the (nonequilibrium) state of the solid. In this case it is necessary to consider the gas and solid as a single nonequilibrium system.

We shall consider the case when  $\omega_0\tau_p \gg 1$  and the absorption of sound in a solid is regarded as the absorption of single acoustic phonons.<sup>4</sup> We can assume<sup>5</sup> that, in spite of the large acoustic wavelength, the scattering of molecules on the surface of a solid is also accompanied by creation and annihilation of acoustic phonons, as well as of phonons of higher frequencies. Within the framework of this mechanism the description of the absorption of sound by a gas clearly requires the development of a kinetic theory of a single nonequilibrium gas-solid system. Such a theory is developed below and it provides a unified description of the attenuation of high-frequency surface sound and of the associated transport processes both in the gas and solid. Our theory will allow us, in particular, to explain the mass and frequency dependences of  $\gamma_2$  observed in the gigahertz range of frequencies,<sup>2</sup> which are known to be due to the competition between the high-frequency mechanism of creation (annihilation) of acoustic phonons by gas molecules and the low-frequency mechanism of the scattering of molecules on

a vibrating surface which is in a local equilibrium. An important feature of the proposed high-frequency mechanism is the drag of molecules by a flux of acoustic phonons which results in macroscopic motion of the gas along the surface. As shown below, under typical experimental conditions the flow velocity may reach an easily measurable value of  $\sim 0.1$  m/sec. We shall use our kinetic theory to predict the frequency and temperature dependences of the coefficient  $\gamma_1$  representing the absorption of surface acoustic waves in a solid. They agree with the available experimental data<sup>6</sup> and with the theoretical dependences obtained earlier<sup>7</sup> for a simple model of a solid by calculating the dynamic matrix of a semi-infinite anharmonic crystal.

In the next section we shall formulate for the distribution functions of surface phonons, bulk phonons, and gas molecules the transport equations and the boundary conditions which allow us to describe the transport processes in a gas-solid system, including those during propagation of high-frequency sound. We shall next consider the attenuation of surface acoustic waves due to the absorption of acoustic phonons by thermal phonons (Sec. 3) and also due to the absorption of acoustic phonons by gas molecules (Sec. 4). In Sec. 5 we shall discuss the flow of a gas due to the drag of molecules by acoustic phonons.

## 2. SYSTEM OF TRANSPORT EQUATIONS AND BOUNDARY CONDITIONS FOR THE DISTRIBUTION FUNCTIONS OF MOLECULES AND PHONONS

We shall consider a system formed by a gas and an insulating single crystal, which are separated by a cleaved surface of the crystal coinciding with the  $z = 0$  plane. We shall distinguish between the subsystems of gas molecules, volume (bulk) phonons, and surface phonons.<sup>8</sup> The subsystem of molecules, in which we shall include those outside the range of action of the surface forces, will be described by a distribution function  $f(\mathbf{v})$ , where  $\mathbf{v}$  is the velocity of a molecule. In the subsystem of volume phonons we shall include those phonons which are localized outside the surface region where the vibrations of atoms of a semi-infinite crystal differ considerably from the vibrations of atoms in an ideal infinite crystal. We shall describe the subsystem of volume phonons by a distribution function  $n(\mathbf{k}, j)$ , which represents the average occupation number of a vibrational mode of an infinite crystal characterized by a wave vector  $\mathbf{k}$  and a polarization  $j$ . The distribution functions  $f$  and  $n$  satisfy the familiar transport equations<sup>4,9</sup>:

$$\frac{\partial f(\mathbf{r}, \mathbf{v}, t)}{\partial t} + \mathbf{v} \frac{\partial f(\mathbf{r}, \mathbf{v}, t)}{\partial \mathbf{r}} = I_M(\{f\}), \quad z > 0, \quad (2.1)$$

$$\frac{\partial n(\mathbf{r}, \mathbf{k}, j, t)}{\partial t} + \mathbf{c}_j \frac{\partial n(\mathbf{r}, \mathbf{k}, j, t)}{\partial \mathbf{r}} = I_V(\{n\}), \quad z < 0. \quad (2.2)$$

Here,  $I_M$  and  $I_V$  are the collision integrals of molecules and phonons, respectively;  $\mathbf{r} = (x, y, z)$  is the spatial coordinate;  $t$  is time;  $\mathbf{c}_j = \partial \omega(\mathbf{k}, j) / \partial \mathbf{k}$ , where  $\omega(\mathbf{k}, j)$  is the frequency of a vibrational mode  $(\mathbf{k}, j)$ .

Near the boundary in a solid there is also the subsystem of surface phonons localized in surface vibrational modes<sup>10,11</sup> of a semiinfinite crystal. The interaction of surface phonons with one another and with volume phonons scattered on the surface is due to the anharmonicity of the atomic vibrations in a solid. It has been shown experimen-

ly<sup>12</sup> that a magnitude of this anharmonicity near the surface is comparable with the anharmonicity of vibrations of atoms in the bulk. Consequently, the anharmonicity in the interaction of phonons near the surface can be regarded as a perturbation in the same sense as the anharmonicity in the bulk of a solid when calculations are made<sup>4,9</sup> of the collision integral  $I_V$  in Eq. (2.2). The interaction of the subsystem of surface phonons with gas molecules, due to the scattering of the latter by the surface of a solid, may also be regarded as weak because the densities of a gas and a solid differ by many orders of magnitude. Then, the transport equation for the distribution function  $\nu$  of surface phonons, representing the average occupation number of each surface mode, can be written in the form

$$\frac{\partial \nu(\mathbf{R}, \mathbf{K}, \xi, t)}{\partial t} + \mathbf{c}_\xi \frac{\partial \nu(\mathbf{R}, \mathbf{K}, \xi, t)}{\partial \mathbf{R}} = I_S(\{f, n, \nu\}), \quad z = 0. \quad (2.3)$$

Here, the collision integral  $I_S$  allows for the interaction of surface phonons with one another and with volume phonons because of the anharmonicity of vibrations of atoms in a solid, and also the interaction of these phonons with gas molecules;  $\mathbf{R} = (x, y)$  is the vector in the  $z = 0$  plane;  $\mathbf{c}_\xi = \partial \omega(\mathbf{K}, \xi) / \partial \mathbf{K}$ , where  $\omega(\mathbf{K}, \xi)$  is the frequency of a surface mode with a wave vector  $\mathbf{K}$  tangential to the surface and a polarization  $\xi$ . We shall assume that the range of variation of  $\mathbf{K}$  coincides with the range of variation of that component of the wave vector of volume vibrations which is tangential to the surface, i.e.,  $\mathbf{k} = (\mathbf{K}, k_z)$ . This can be ensured by a suitable selection of a unit cell of the crystal lattice in the interior of the solid. The transport equation (2.3) applies to phonons, including high-frequency acoustic phonons, whose frequencies satisfy the inequality  $\omega \tau_p \gg 1$ . This is equivalent to  $L \ll \lambda_p$ , where  $L \sim c/\omega$  is the size of the region where the surface has a significant influence on the vibrations of atoms in a solid ( $c$  is the velocity of sound in the solid) and  $\lambda_p = c\tau_p$  is the mean free path of typical thermal phonons. This allows us to regard surface phonons as "two-dimensional" and localized in the  $z = 0$  plane within the framework of the kinetic theory.

We shall find  $I_S$  of Eq. (2.3) by considering first the boundary between a solid and vacuum. In this case the change in the occupation numbers of vibrational modes of a semi-infinite crystal is due to the anharmonic part of the potential energy of the interaction of atoms in a solid. The first anharmonic (cubic) term in the expansion of the potential energy is of the form

$$H^{(3)} = \frac{1}{6} \sum_{\substack{n, s}} \Lambda_{\beta_1 \beta_2 \beta_3 n_1 n_2 n_3}^{s_1 s_2 s_3} U_{s_1 n_1 \beta_1} U_{s_2 n_2 \beta_2} U_{s_3 n_3 \beta_3}. \quad (2.4)$$

Here,  $\mathbf{U}_{sn}$  are the displacement vectors of atoms in the lattice;  $\beta_1, \beta_2$ , and  $\beta_3$  are the indices which assume the values  $x, y$ , and  $z$ ;  $s_1, s_2$ , and  $s_3$  are the numbers of atoms in a unit cell; the index  $n$  includes integers  $l_1$  and  $l_2$  describing the position of a unit cell in the plane tangential to the surface, as well as an integer denoting the layer number  $l_3$ ; the summation is carried out over all values of  $n$  and all  $s$ .

The displacement  $\mathbf{U}_{sn}$  is determined by a superposition of normal vibrations of a semiinfinite crystal. The set of these vibrations includes surface modes  $(\mathbf{K}, \xi)$  as well as volume modes, which are defined uniquely by specifying the asymptotic behavior in the interior of a crystal. We shall select

them in such a way that each mode corresponds to a wave incident on the surface and characterized by a wave vector  $\mathbf{k}$ , a polarization  $j$ , a frequency  $\omega(\mathbf{k}j)$ , a group velocity component  $c_z(\mathbf{k}j) = \partial\omega(\mathbf{k}j)/\partial k_z \geq 0$  normal to the surface and a corresponding set of reflected waves. This definition of volume modes corresponds to the representation of diverging waves.<sup>13</sup> In such a representation the operator describing the displacement of a lattice atom  $\hat{U}_{sn}$  can be described by

$$\begin{aligned} \hat{U}_{sn} = & \sum_{\mathbf{k}j} \left[ \frac{\hbar}{2MN\omega(\mathbf{k}j)} \right]^{1/2} [e(l_{3s}; \mathbf{k}j) \exp(i\mathbf{K}\mathbf{r}_n) \hat{a}_{\mathbf{k}j} \\ & + e^*(l_{3s}; \mathbf{k}j) \exp(-i\mathbf{K}\mathbf{r}_n) \hat{a}_{\mathbf{k}j}^+] + \sum_{\mathbf{K}\xi} \left[ \frac{\hbar}{2MN_\tau\omega(\mathbf{K}\xi)} \right]^{1/2} \\ & \times [e(l_{3s}; \mathbf{K}\xi) \exp(i\mathbf{K}\mathbf{r}_n) \hat{a}_{\mathbf{K}\xi} + e^*(l_{3s}; \mathbf{K}\xi) \exp(-i\mathbf{K}\mathbf{r}_n) \hat{a}_{\mathbf{K}\xi}^+]. \end{aligned} \quad (2.5)$$

The summation is carried out over all volume and surface modes;  $\hat{a}^+$  and  $\hat{a}$  are the phonon creation and annihilation operators;  $\mathbf{r}_n$  is the vector governing the equilibrium position of a unit cell  $n$ ;  $M$  is the mass of this unit cell;  $N$  and  $N_\tau$  are the numbers of unit cells in the solid and in one layer, respectively; in the limit  $l_s \rightarrow \infty$  the vector  $\mathbf{e}$  tends to zero for a surface mode and it represents a superposition of the polarization vectors of waves incident on and reflected from the surface in the case of a volume mode.

Substitution of the displacement operators  $\mathbf{U}$  of Eq. (2.5) into Eq. (2.4) gives, in place of the vectors  $\mathbf{U}$ , an operator  $\hat{H}^{(3)}$  which contains  $\hat{a}$  and  $\hat{a}^+$  in various combinations of three at a time. The annihilation of a surface phonon is assumed to occur in seven possible processes: decay of a surface phonon into two surface phonons, two volume phonons, or one surface and one volume phonon; coalescence of a surface phonon with another surface phonon to form a volume or a surface phonon; coalescence of a surface phonon with a volume phonon to form a surface or a volume phonon. Creation of a surface phonon occurs in processes which are opposite to those just listed. If we regard  $\hat{H}^{(3)}$  as a perturbation, then by analogy with  $I_V$  of Eq. (2.2) (Refs. 4 and 9) we can readily find the part of the collision integral of surface phonons which is associated with the anharmonicity and is described by

$$I_{S1}(\mathbf{K}\xi) = \sum_i I_{S1}^{(i)}(\mathbf{K}\xi). \quad (2.6)$$

The summation is carried out over all seven collision processes described above and each term  $I_{S1}^{(i)}$  represents the difference in the average number of phonons created and annihilated in a mode  $(\mathbf{K}, \xi)$  as a result of a collision of this type. We shall give the explicit form of the term  $I_{S1}^{(1)}$  describing the process of coalescence of a surface phonon with a volume phonon producing a volume phonon and the reverse process:

$$I_{S1}^{(1)}(\mathbf{K}\xi) = \sum_{\mathbf{k}_1j_1(c_{z1}>0)} \sum_{\mathbf{k}_2j_2(c_{z2}>0)} w_{j_1j_2\xi}(\mathbf{K}, \mathbf{k}_1; \mathbf{k}_2) \delta(\omega_2 - \omega_1 - \omega) \times [(v_1+1)(n_1^++1)n_2^+ - v n_1^+(n_2^++1)], \quad (2.7)$$

$$w_{j_1j_2\xi}(\mathbf{K}, \mathbf{k}_1; \mathbf{k}_2) = \frac{\pi\hbar}{2MNN_z\omega_1\omega_2} |\Omega_{j_1j_2\xi}(\mathbf{K}, \mathbf{k}_1; \mathbf{k}_2)|^2, \quad (2.8)$$

$$\Omega_{j_1j_2\xi}(\mathbf{K}, \mathbf{k}_1; \mathbf{k}_2) = \frac{1}{MN_\tau} \sum_{(n,s)} \Lambda_{\beta_1\beta_2\beta_3n_1n_2s} e_{\beta_1}(l_{3s}; \mathbf{K}\xi)$$

$$\times e_{\beta_2}(l_{3s_1}; \mathbf{k}_1j_1) e_{\beta_3}^*(l_{3s_2}; \mathbf{k}_2j_2) \exp[i(\mathbf{K}\mathbf{r}_n + \mathbf{K}_1\mathbf{r}_{n_1} - \mathbf{K}_2\mathbf{r}_{n_2})]. \quad (2.9)$$

Here,

$$\begin{aligned} \omega = \omega(\mathbf{K}\xi), \quad \omega_1 = \omega_1(\mathbf{k}_1j_1), \quad \omega_2 = \omega_2(\mathbf{k}_2j_2), \\ v = v(\mathbf{K}\xi), \quad n_1^+ = n_1^+(\mathbf{k}_1j_1), \quad n_2^+ = n_2^+(\mathbf{k}_2j_2), \quad N_z = N/N_\tau. \end{aligned}$$

In writing down Eq. (2.7) we have to allow for the fact that the average occupation numbers of volume modes in the adopted representation of diverging waves represent the distribution function  $n^+$  of volume phonons incident on the surface and deduced from the transport equation (2.2). It should be pointed out that because of periodicity of a crystal in the direction tangential to the surface the value of  $\Omega$  of Eq. (2.9) differs from zero only if  $\mathbf{K} + \mathbf{K}_1 - \mathbf{K}_2 = \mathbf{b}$ , where  $\mathbf{b}$  is the reciprocal lattice vector which is tangential to the surface. The other terms on the right-hand side of Eq. (2.6) are similar to Eqs. (2.7)–(2.9).

The influence of gas molecules and the distribution function  $\nu$  of surface phonons is due to the possibility of creation (annihilation) of single phonons by molecules. We shall assume that the average time  $t_a$  which a molecule spends in the region where the surface forces are acting (the size of the region being  $r_0 \sim 10^{-8}$  cm) is much less than the characteristic time  $t_p$  of decay of the vibrational state of a solid. In particular, when molecules are scattered without being captured by the surface, we have  $t_a \sim r_0(2T/m)^{-1/2} \sim 10^{-12}$  sec, and the role of  $t_p$  is played by the lifetime of thermal phonons  $\tau_p \sim 10^{-10} - 10^{-11}$  sec. Then, we can ignore the phonon-phonon interaction and the process of scattering of molecules on the surface. It follows that the change in the number of phonons in the surface mode because of the phonon-phonon interaction and the interaction with gas molecules occur independently and, consequently, the corresponding processes can make additive contributions to  $I_S$ :

$$I_S(\mathbf{K}\xi) = I_{S1}(\mathbf{K}\xi) + I_{S2}(\mathbf{K}\xi). \quad (2.10)$$

Here,  $I_{S2}$  is the average change in the number of phonons in a surface mode  $(\mathbf{K}, \xi)$  per unit time due to the scattering of molecules on the surface. We can determine  $I_{S2}$  by noting that if  $t_a \ll t_p$ , then collisions of molecules with the surface can be discussed in terms of the scattering theory and the probability  $W_D$  of a transition in a semiinfinite crystal from a state  $p'$  [the state  $p' \equiv \{N'_{kj}\}\{N'_{\mathbf{K}\xi}\}$  is characterized by a set of initial occupation numbers of volume ( $N_{\mathbf{K}j}$ ) and surface ( $N'_{\mathbf{K}\xi}$ ) vibrational modes of a semi-infinite crystal] to a state  $p$  as a result of a transition of a molecule from a state with a velocity  $\mathbf{v}'$  (Refs. 13 and 14):

$$W_D(\mathbf{v}'p' \rightarrow \mathbf{v}p) = \frac{2\pi}{\hbar|v_z'|} \left( \frac{m}{2\pi\hbar} \right)^3 |(v p | \hat{T} | v' p')|^2 \delta(\epsilon - \epsilon'). \quad (2.11)$$

Here,  $\hat{T}$  is the operator of the transition governed by the potential of the molecule-surface interaction;  $\epsilon'$  and  $\epsilon$  represent the total energy of the system before and after a collision. Using Eq. (2.11) and calculating the average change in the number of phonons in a fixed surface mode  $(\mathbf{K}, \xi)$ , we can represent the expression for  $I_{S2}$  as follows:

$$I_{S2}(\mathbf{K}\xi) = S \int_{v_z>0} d\mathbf{v} \int_{v_z<0} d\mathbf{v}' |v_z'| f^-(\mathbf{v}') [(v+1)W^+(\mathbf{v}' \rightarrow \mathbf{v}; \mathbf{K}\xi) - vW^-(\mathbf{v}' \rightarrow \mathbf{v}; \mathbf{K}\xi)]. \quad (2.12)$$

Here,  $f^-$  is the distribution function of molecules incident on the surface, deduced by solving the transport equation (2.1),

$S$  is the surface area, and  $W^\pm$  are the probabilities of creation and annihilation of a phonon  $(\mathbf{K}, \xi)$  when molecules are scattered by the surface. In general, when an arbitrary number of other phonons is interchanged between a molecule and a solid, the probabilities  $W^\pm$  depend on the average occupation numbers of the corresponding vibrational modes. It should be stressed that the dependence of  $I_{S_2}$  and  $\nu$  on  $f^-$  [see Eq. (2.3)] follows from Eq. (2.12).

The system of transport Eqs. (2.1)–(2.3) is closed as a result of formulation of the boundary conditions for  $f$  and  $n$  on the  $z = 0$  surface. In general, the distribution functions of gas molecules incident  $f^-$  on and reflected  $f^+$  from the surface are related by<sup>4</sup>

$$|v_z|f^+(\mathbf{v}) = \int_{v'_z < 0} dv' |v'_z| f^-(\mathbf{v}') W(\mathbf{v}' \rightarrow \mathbf{v}). \quad (2.13)$$

Here, the kernel  $W$  represents the probability density of a transition of a molecule from a state with a velocity  $\mathbf{v}'$  to a state with a velocity  $\mathbf{v}$  when the molecule is scattered by the surface and it can be expressed in a familiar manner in terms of the probability  $W_D$  of Eq. (2.11) by averaging it over the initial states and summing over the final states of a solid.<sup>15,16,1</sup> Then, the value of  $W$  (see Ref. 1) depends on the distribution functions  $n^+$  and  $\nu$  satisfying Eqs. (2.2) and (2.3).

The boundary condition relating the distribution functions of phonons incident  $n^+$  and reflected  $n^-$  by the surface can be formulated if we allow for surface modes and for the phonon-phonon interaction near the surface in the boundary condition obtained in Ref. 1. This gives

$$\frac{|c_z|}{L_z} n^-(\mathbf{k}j) = \sum_{\mathbf{k}_1 j_1 (c_{z1} > 0)} \left[ \frac{c_{z1}}{L_z} n^+(\mathbf{k}_1 j_1) + \dot{N}_M(\mathbf{k}_1 j_1) + \dot{N}_p(\mathbf{k}_1 j_1) \right] V_p(\mathbf{k}_1 j_1 \rightarrow \mathbf{k}j; \omega). \quad (2.14)$$

Here,  $V_p$  is the probability of a transition of a phonon  $\hbar\omega$  from a state  $(\mathbf{k}_1, j_1)$  to a state  $(\mathbf{k}j)$  when the phonon is scattered by the surface;  $L_z$  is the thickness of the solid; the quantities  $\dot{N}_M$  and  $\dot{N}_p$  represent the average change in the number of phonons in a mode  $(\mathbf{k}_1, j_1)$  per unit time, which is due to the scattering of molecules on the surface and due to collisions of phonons in the surface region. The quantity  $\dot{N}_M$  is given by an expression of the (2.12) type where the distribution function  $\nu$  is replaced with the distribution function  $n_1^+$ . The quantity  $\dot{N}_p$  is obtained from the same considerations as  $I_{S_1}$  and it has seven terms corresponding to different types of phonon-phonon collisions which are not given here because they are too cumbersome.

The transport Eqs. (2.1)–(2.3) and the boundary conditions (2.13) and (2.14) represent the required system of equations for the description of the transport processes in the nonequilibrium gas-solid system including the processes occurring in the case of propagation of high-frequency sound in a solid. (In the latter case in addition to the condition  $\omega_0 \tau_p \gg 1$  we must clearly satisfy also the condition  $\omega_0 \tau_M \gg 1$ , where  $\tau_M^{-1}$  is the frequency of intermolecular collisions in the gas. This condition ensures the absence of an acoustic wave in the gas and for  $\omega_0 \sim 10^{10}$  Hz it is satisfied right up to gas pressures of tens of atmospheres.) In particular, the energy fluxes in the solid ( $Q_s$ ) and in the gas ( $Q_g$ ) due to the attenuation of surface acoustic waves can be deter-

mined by two equivalent methods. In the first we calculate the distribution functions  $f$  and  $n$ , which after suitable integration give the expressions for  $Q_g$  and  $Q_s$ . In the second case used below (see Secs. 3 and 4) we shall consider each of the terms in  $I_S$  of Eq. (2.10) and these are clearly related to  $Q_s$  and  $Q_g$  by the factor  $\hbar\omega_0$ . It is then convenient to analyze the quantities which are usually determined experimentally and which are related linearly to the macroscopic energy fluxes  $Q_s$  and  $Q_g$ : these are the terms  $\gamma_1$  and  $\gamma_2$  of the absorption coefficient of surface acoustic waves ( $\gamma = \gamma_1 + \gamma_2$ ), due to the interaction of acoustic phonons with thermal phonons ( $\gamma_1 = Q_s/R$ , where  $R$  is the acoustic energy density) and with gas molecules ( $\gamma_2 = Q_g/R$ ).

### 3. ABSORPTION OF SURFACE SOUND BY VOLUME PHONONS

We shall consider the propagation of a high-frequency surface acoustic wave along the boundary of a solid coinciding with the  $z = 0$  plane in the absence of a gas above the surface. Using the transport Eq. (2.3), we shall determine the dependence of  $\gamma_2$  on  $\omega_0$  and  $T$ . We shall assume that the phonon frequencies are linear functions of the wave vectors. We can then easily show that the laws of conservation of energy and the quasimomentum tangential to the surface can be satisfied simultaneously in three-phonon processes involving an acoustic phonon only in collisions of an acoustic phonon with a volume phonon producing a volume phonon, as described by Eqs. (2.7)–(2.9). We shall assume that apart from acoustic phonons, the other phonons participating in such collisions have an equilibrium distribution:  $n_1^+ = n_{1B}$  and  $n_2^+ = n_{2B}$ , where  $n_B$  is the Bose-Einstein distribution. (A change in the temperature of a solid near the surface associated with the attenuation of sound is small and for values of the acoustic energy density  $R \sim 10$  erg/cm<sup>2</sup> at  $\omega_0 \sim 10^{10}$  Hz and  $T \sim 300$  K it does not exceed 0.1 K if  $\gamma_1 \sim 10^6$  sec<sup>-1</sup>—see Ref. 17.) Then, using Eqs. (2.7)–(2.9) and retaining only the terms of the first order in  $\hbar\omega_0/T$ , we obtain the following expression for  $\gamma_1$  if  $\hbar\omega_0/T \gg 1$ :

$$\gamma_1 = \frac{I_{S_1}^{(1)}(\mathbf{K}_0 \xi_0)}{\nu_0} = \frac{\hbar\omega_0}{T} \sum_{\mathbf{k}_2 j_2 (c_{z2} > 0)} \sum_{\mathbf{k}_1 j_1 (c_{z1} > 0)} \times w_{j_1 j_2 \xi_0}(\mathbf{K}_0, \mathbf{k}_1; \mathbf{k}_2) n_{1B} (n_{1B} + 1) \delta(\omega_2 - \omega_1 - \omega_0). \quad (3.1)$$

Here,  $\nu_0 = \nu_0(\mathbf{K}_0 \xi_0) \gg 1$  is the average occupation number of an acoustic mode  $(\mathbf{K}_0, \xi_0)$  and  $\mathbf{K}_2 = \mathbf{K}_1 + \mathbf{K}_0$ .

We shall calculate the frequency dependence of  $\gamma_1$  by determining the dependence of  $w$  on  $K_0$ . If we consider an acoustic wave in terms of the theory of elasticity,<sup>11</sup> when  $|e(I_{3S}; \mathbf{K}_0 \xi_0)| \propto K_0^{1/2}$  (Ref. 18), and apply Eqs. (2.7) and (2.8) to the usual procedures employed in the derivation of the frequency dependence of the absorption coefficient of high-frequency sound in the bulk of a solid,<sup>4,9</sup> we can show that  $w$  is independent of  $\mathbf{K}_0$ , but depends only on its direction. [It should be pointed out that if long-wavelength volume phonons participate in the three-phonon process of Eq. (3.1), we can similarly obtain the dependence  $w \propto k_1 k_2$ .] Therefore, the absorption coefficient of high-frequency surface acoustic waves is found to be proportional to  $\omega_0$  for any value of  $T$ , exactly as in the absorption of sound in the bulk of a solid,<sup>4</sup> and this is due to the interaction of acoustic phonons with thermal phonons in accordance with Eq. (3.1).

The temperature dependence of  $\gamma_1$  observed at low tem-

peratures when  $T \ll \Theta$  ( $\Theta$  is the Debye temperature) can be explained by noting that the main contribution to Eq. (3.1) comes from phonons with the wave vectors  $k_1, k_2 \sim T/c_j \hbar \ll a^{-1}$ , where  $a$  is the characteristic size of a unit cell in the investigated solid. Then, all the three phonons participating in a collision can be regarded as long-wavelength phonons and we can use the linear dependence of  $w$  on  $k_1$  and  $k_2$ . Summing in Eq. (3.1) with respect to  $k_{2z}$ , we obtain

$$\gamma_1 = \frac{\hbar \omega_0}{T} \sum_{j,j_2} \int_{(c_{j_2} > 0)} dk_1 \frac{1}{(2\pi)^3} A k_1^2 \frac{c_{j_1}}{c_{j_2}^2} n_{1B} (n_{1B} + 1). \quad (3.2)$$

Here, the value of  $A$  depends only on the orientation of the vectors  $\mathbf{K}_0$  and  $\mathbf{k}_1$ . Extending formally integration with respect to  $k_1$  to infinity, we obtain

$$\gamma_1 \propto \omega_0 T^4. \quad (3.3)$$

Such a dependence has been observed experimentally<sup>6</sup> at  $\omega_0 = 10^{19}$  Hz and  $T \sim 60$  K. The frequency and temperature dependences of the type given by Eq. (3.3) were obtained earlier<sup>7</sup> using a simple model of the lattice and the dynamic matrix of a semi-infinite anharmonic signal.

If  $\Theta \ll T$ , then Eq. (3.1) is dominated by the values of  $k_1$  and  $k_2 \propto \Theta/c_j \hbar \propto a^{-1}$ . It then follows from Eqs. (2.8) and (2.9) that the value of  $w$ , like the corresponding probability of the phonon-phonon interaction in the interior of a solid,<sup>9</sup> can be regarded as independent of the wave vectors of thermal phonons. Ignoring the contribution of optical phonons to  $\gamma_1$  and integrating Eq. (3.1), we obtain

$$\gamma_1 \propto \omega_0 T. \quad (3.4)$$

The dependences (3.3) and (3.4) are similar to the corresponding dependences of the absorption coefficient of transverse sound in the bulk of a solid.<sup>4,9</sup>

#### 4. ABSORPTION OF SURFACE SOUND BY GAS MOLECULES

We shall calculate the term  $\gamma_2$  in the absorption coefficient of surface acoustic waves by using the one-phonon approximation developed in Ref. 19 for the calculation of the transition probability  $W_D$  of Eq. (2.11), which is used to allow for direct transitions from the initial state of the molecule-solid system to the final state, as well as for transitions accompanied by the creation and annihilation of a virtual phonon on condition that the total energy of the system is conserved. In contrast to the simpler first Born approximation in the method of distorted waves,<sup>13</sup> which allows only for direct transitions in the system, this approximation makes it possible to obtain the probability  $W_D$  for molecules of arbitrary mass<sup>19,14</sup> normalized to unity and suitable, for example, in the analysis of the dependence of  $\gamma_2$  on the mass of molecules  $m$  in a wide range of masses.<sup>11</sup> In this approximation the probabilities of creation  $W^+$  and annihilation  $W^-$  of an acoustic phonon [see Eq. (2.12)] can be obtained in the form

$$W^\pm(\mathbf{v}' \rightarrow \mathbf{v}; \mathbf{K}_0 \xi_0) = P^\pm(\mathbf{v}' \rightarrow \mathbf{v}; \mathbf{K}_0 \xi_0) [1 + {}^{1/2} P_\Sigma(\mathbf{v}')]^{-2}, \quad (4.1)$$

$$P^\pm(\mathbf{v}' \rightarrow \mathbf{v}; \mathbf{K}_0 \xi_0) = \frac{2\pi}{\hbar |v_z'|} \left( \frac{m}{2\pi\hbar} \right)^3$$

$$\times \delta \left( \frac{mv^2}{2} \pm \hbar\omega_0 - \frac{mv'^2}{2} \right) |\lambda| \hbar^\pm |\lambda'|^2. \quad (4.2)$$

Here,  $P^+$ ,  $P^-$ , and  $P_\Sigma$  are, respectively, the probabilities of

inelastic scattering of molecules accompanied by creation and annihilation of an acoustic phonon (accurate to within a factor  $\nu_0$ ) and the total probability of nonspecular reflection in the first Born approximation of the method of distorted waves.<sup>13,14</sup> The unperturbed potential  $V_0$  is selected to be that part of the potential of the interaction of a molecule with a frozen lattice which depends only on the distance of the molecule from the surface and the perturbation  $V_1$  is associated with thermal vibrations of the lattice. We can then show<sup>1</sup> that the operator  $h^-$  is given by  $h^- = \exp(i\mathbf{K}_0 \mathbf{r}) \tilde{V}$  and that  $h^+ = h^{-*}$  (here,  $\mathbf{r}$  is the coordinate of the molecule and  $\tilde{V}$  depends only on the distance between the molecule and the surface), and the wave function  $\lambda$ ) determines the state of the molecule in the potential  $V_0$  and is a solution of the corresponding Schrodinger equation. The probability  $P_\Sigma$  can be calculated by summing the probabilities of an inelastic interaction of a molecule with each mode of a solid; the structure of these probabilities is similar to that described by Eq. (4.2).

We shall now discuss the model of a solid in the form of a quasicontinuum with a free boundary<sup>11</sup> and we shall quantize the vibrational spectrum in the same way as in Ref. 18. Assuming that the equilibrium position of the boundary of the continuum coincides with the  $z = 0$  plane, we shall select the potential  $V$  in the form

$$V = D \exp \{-2q^{-1} [z - \hat{U}_z(x, y, 0)]\}. \quad (4.3)$$

Here,  $D$  and  $q$  are, respectively, the characteristic energy and the radius of action of the surface forces;  $x, y$ , and  $z$  are the coordinates of a molecule;  $\hat{U}_z$  is the  $z$  component of the operator representing the displacement of an element of the surface of the continuum with the equilibrium coordinates  $(x, y, 0)$  as a result of thermal vibrations. The nature of  $\hat{U}$  is given by Eq. (2.5) where the vectors  $\mathbf{r}_n$  and  $\mathbf{e}$  should be regarded as continuous functions of the spatial coordinates.

Equations (2.12) and (4.1)–(4.3) allow us to calculate  $\gamma_2$  if we know the distribution functions  $n^+$ ,  $\nu$ , and  $f$ . Ignoring small changes in the temperature of a solid associated with the absorption of sound (see Sec. 3), we shall assume that  $n^+ = n_B$  and  $\nu = \nu_B$  for all the modes except the acoustic one. We shall also bear in mind that in the range of linear absorption of sound by a gas, which we are considering here, the average change in the energy of a molecule due to nonequilibrium occupancy of an acoustic mode does not exceed  $\hbar\omega_0$  and is much smaller than its thermal energy. Consequently, the difference between the distribution function  $f$  and its equilibrium form  $f_M$  ( $f_M$  is the Maxwellian distribution with an equilibrium density of the gas molecules  $\tilde{n}_M$  and a temperature  $T$ ) is small and in the determination of  $\gamma_2$  we can substitute  $f^- = f_M$  in Eq. (2.12). Then, for an infinitely high potential barrier [ $q \rightarrow 0$ , see Eq. (4.3)] we obtain the following expression for  $\gamma_2$ :

$$\gamma_2 = \tilde{n}_M \omega_0 \left( \frac{2mT}{\pi} \right)^{3/2} \frac{\Phi}{\rho_s c_0}$$

$$\times \int_0^\infty du u e^{-u} \left[ 1 + \frac{P_\Sigma(u)}{4} \right]^{-2} \left[ \left( 1 + \frac{P_\Sigma(u)}{4} \right)^{-1} \right.$$

$$\left. \times \frac{1}{2} \frac{\partial P_\Sigma}{\partial u} + 1 \right], \quad (4.4)$$

$$P_z(u) = \frac{mT^3 F}{\pi^2 \rho_s \hbar^3 c_t^3} \left\{ \int_0^{\tau(u)} d\epsilon \epsilon [u(u-\epsilon)]^{1/2} [n_B(\epsilon) + 1] + \int_0^{e/\tau} d\epsilon \epsilon [u(u+\epsilon)]^{1/2} n_B(\epsilon) \right\}, \quad (4.5)$$

$$\Phi = \frac{1}{4} \xi^4 \left[ \frac{2-\xi^2}{2(1-\xi^2)^{1/2}} + \frac{2-\eta^2 \xi^2}{2(1-\eta^2 \xi^2)^{1/2}} \left( \frac{1-\xi^2}{1-\eta^2 \xi^2} \right)^{1/2} - \frac{2-\xi^2}{(1-\eta^2 \xi^2)^{1/2}} \right]^{-1} \quad (4.6)$$

Here,  $u$  is the dimensionless energy of the motion of a molecule in the direction normal to the surface;  $\epsilon$  is the dimensionless energy of a phonon created or annihilated by the scattering of a molecule;  $\tau(u) = \min\{\Theta/T, u\}$ ;  $c_0$  is the velocity of surface acoustic waves;  $\rho_s$  is the density of the investigated solid;  $\eta = c_t/c_l$ ;  $\xi = c_0/c_l$ ;  $c_t$  and  $c_l$  are the velocities of transverse and longitudinal sound in the bulk of the solid; the quantity  $F$  depends only on  $\eta$  and it is described by Eq. (2.12) of Ref. 18.

It follows from Eq. (4.4) that  $\gamma_2$  depends linearly on the gas density and on the frequency of surface acoustic waves. This is in qualitative agreement with the results obtained both within the framework of the theory of propagation of waves at the boundary of two continuous media<sup>17</sup> and from the description of a gas bounded by a local-equilibrium vibrating surface based on the kinetic theory [see Eq. (4.7) below]. In an analysis of the mass and temperature dependences of  $\gamma_2$  we note, as is easily demonstrated on the basis of Eq. (4.5), that in the limit of low values of  $m$  and  $T$  we have  $P_z \ll 1$ . We then obtain  $\gamma_2 \propto (mT)^{1/2}$ . As  $m$  or  $T$  increases, the dependence of  $\gamma_2$  on the mass of molecules and temperature becomes weaker and disappears in the case of large values of  $m$  or  $T$  (Fig. 1) This is a basic feature of the mechanism of the absorption of acoustic phonons by molecules since in the interaction of two continuous media when molecules are scattered by a local-equilibrium vibrating surface we have  $\gamma_2 \propto (mT)^{1/2}$  for any value of  $m$  and  $T$  [see Ref. 17 and Eq. (4.7)].

Although Eq. (4.4) was derived using a number of sim-

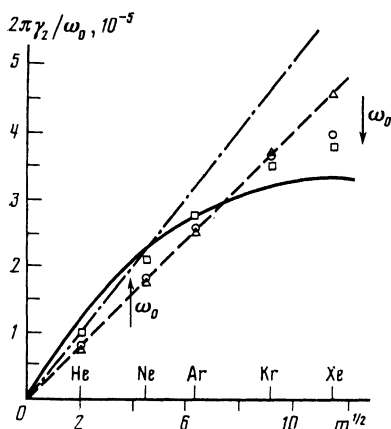


FIG. 1. Dependence of  $\gamma_2$  on the mass of gas molecules  $m$ . The continuous curve represents the dependence of Eq. (4.4) and the dashed curve represents Eq. (4.7). The chain line corresponds to the theory<sup>17</sup> of the propagation of sound at the boundary between two continuous media. The experimental points are taken from Ref. 2:  $\Delta$ )  $\omega_0/2\pi = 0.54 \times 10^9$  Hz;  $\square$ )  $1.42 \times 10^9$  Hz;  $\square$ )  $2.43 \times 10^9$  Hz.

ple models, the qualitative conclusions relating to the dependences of  $\gamma_2$  on  $\omega_0$ ,  $m$ , and  $T$  are clearly of general validity. In fact, returning to the general formula (2.12) we note that in the region of linear absorption of sound by a gas the expression in the square brackets on the right-hand side reduces to a difference between the total probability of the scattering of molecules accompanied by the absorption of an acoustic phonon and its creation (for given values of  $\nu^1$  and  $\nu$  and exchange of an arbitrary number of other phonons). Integrating Eq. (2.12) with respect to the velocities and expanding the probabilities in powers of a small quantity  $\hbar\omega_0/T$ , we find that zeroth terms of this expansion are equal because creation or annihilation of a phonon with zero energy cannot alter the probability of the scattering of a molecule. When an allowance is made for first-order nonvanishing terms, we can obtain  $\gamma_2 \propto \omega_0$ . In an analysis of the mass dependence of  $\gamma_2$  we shall use the familiar result<sup>14</sup> that in the case of light molecules the probability of inelastic scattering and, consequently,  $W^\pm$  in Eq. (2.12) is proportional to  $m/M_s$ , where  $M_s$  is the mass of atoms in the solid. Bearing in mind that the flux of molecules incident on a surface is proportional to  $m^{-1/2}$  we obtain  $\gamma_2 \propto m^{1/2}$ . However, since the probability of inelastic scattering  $W_D$  is normalized, its linear dependence on  $m$  cannot be conserved for any value of  $m$ . Consequently, we can expect an increase in  $m$  to make the dependence of  $\gamma_2$  on  $m$  weaker than  $\gamma_2 \propto m^{1/2}$ . Similar reasoning can be applied also to the temperature dependence of  $\gamma_2$ .

To the best of our knowledge, experimental data on the influence of a gas on the absorption of surface sound are available only for frequencies not exceeding several gigahertz<sup>2,17</sup> when  $\omega_0 \tau_p \sim 1$ . Therefore, in an analysis of the experimental dependences of  $\gamma_2$  on  $m$ ,  $\omega_0$ , and  $\tilde{n}_M$  (Ref. 2), we find that in addition to the results of Eqs. (4.4)–(4.6), obtained in the limiting case of high frequencies  $\omega_0 \tau_p \gg 1$ , it is necessary to know the analogous dependences in the limiting case of low frequencies  $\omega_0 \tau_p \ll 1$ . In the latter case we can assume that the molecules are scattered by a local-equilibrium element of the surface experiencing macroscopic vibrations during propagation of sound (see also Ref. 20). In the case of full accommodation of molecules on the surface the energy flux  $Q_g$  which appears in the gas is clearly proportional to the flux of the particles incident on the surface  $\tilde{n}_M (2T/m)^{1/2}$  and to the energy  $mu^2/2$  absorbed by a molecule scattered by an element of the surface vibrating at an average velocity  $u$ . Relating  $u$  and  $\omega_0$  to the formulas of the theory of elasticity<sup>11</sup> and going over from  $Q_g$  to  $\gamma_2$ , we obtain  $\gamma_2 \propto \tilde{n}_M \omega_0 m^{1/2}$ . Omitting cumbersome intermediate steps, we shall simply say that a rigorous kinetic calculation of  $\gamma_2$  carried out by us for the diffuse scattering of molecules on an element of the surface gives

$$\gamma_2 = \frac{Q_g}{R} = \tilde{n}_M \omega_0 \left( \frac{mT}{8\pi} \right)^{1/2} \Phi \frac{1}{\rho_s c_0} [4\xi^{-4} \Omega + 5/2], \quad (4.7)$$

$$\Omega = [(1-\xi^2)^{1/2} - 1/2(2-\xi^2)(1-\eta\xi)^{-1/2}]^2. \quad (4.8)$$

Figures 1 and 2 show the theoretical dependences of  $\gamma_2$  on  $m$  and  $\omega_0$  calculated for the two limiting cases corresponding to the experimental conditions. The continuous curves represent Eq. (4.4) for the high-frequency mechanism of the absorption of acoustic phonons by molecules. The dashed curves represent the dependence (4.7). It is

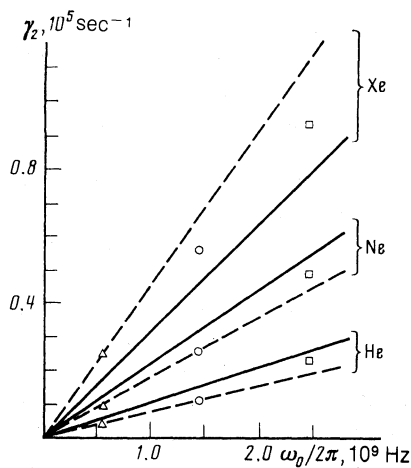


FIG. 2. Dependence of  $\gamma_2$  on the frequency of sound  $\omega_0$ . The notation is the same as in Fig. 1.

clear from Fig. 1 that the low-frequency mechanism can describe the mass dependence of  $\gamma_2$  in the case of the smallest of the frequencies investigated in Ref. 2. However, deviations from the dependence  $\gamma_2 \propto m^{1/2}$  observed both at low and high values of  $m$  on increase in  $\omega_0$  cannot be described by the mechanism of the scattering of molecules on a local-equilibrium vibrating surface element.<sup>2)</sup> On the other hand, the dependence of  $\gamma_2$  on  $m$  deduced on the basis of the mechanism of the absorption of acoustic phonons by gas molecules is in satisfactory qualitative agreement with a dependence obtained at low or high masses at the maximum frequency. The observed frequency dependence  $\gamma_2$  (Fig. 2) is not linear, whereas for both limiting cases  $\omega_0\tau_p \gg 1$  and  $\omega_0\tau_p \ll 1$ , we find that  $\gamma_2 \propto \omega_0$ . However, the mass and the nonlinear frequency dependence can be described if we assume that under experimental conditions there is a competition between the low-frequency mechanism of the interaction of molecules with a vibrating solid in a local equilibrium and the high-frequency mechanism of absorption of acoustic phonons by molecules. Clearly, it follows from these figures that an increase in  $\omega_0$  shifts the experimental points away from the dashed to the continuous curves. Therefore, the deviations from the dependence  $\gamma_2 \propto \omega_0 m^{1/2}$  observed at high frequencies support the high-frequency mechanism of the absorption of acoustic phonons by molecules.

## 5. FLOW OF A GAS DURING PROPAGATION OF A HIGH-FREQUENCY SURFACE ACOUSTIC WAVE

When molecules are scattered on a periodic surface, the change in the tangential component of the momentum is given (to within that component of the reciprocal lattice vector which is tangential to the surface) by the difference between the tangential momenta of annihilated and created phonons. We can therefore assume that the predominant absorption of acoustic phonons by molecules in the course of their scattering on the surface along which a surface acoustic wave is propagating will result in the flow of a gas along the surface.

We shall consider a gas between the surfaces  $z = 0$  and  $z = d$  of two solids and we shall assume that a high-frequency surface acoustic wave is propagating along the  $z = 0$  surface in the positive direction of the  $x$  axis. We consider only the case of a collisionless gas ( $K_n = \lambda_M/d \gg 1$ , where  $\lambda_M$  is the mean free path of molecules in the case of intermolecular collisions), when the transport processes are governed en-

tirely by the interaction of molecules with the surface and the interphase effects would obviously be strongest. Then, in Eq. (2.1) we shall assume (unless otherwise stated) that  $I_M = 0$ . As before, we shall ignore the difference between the average occupation numbers  $n$  and  $\nu$  of all the vibrational modes of the investigated solid (with the exception of the acoustic mode) from the equilibrium values  $n_B$  and  $\nu_B$ , respectively. Therefore, under steady-state conditions the system of transport Eqs. (2.1)–(2.3) reduces, subject to Eq. (3.1) to the following equations for the molecules of a gas and for acoustic phonons:

$$v_x \partial f / \partial x + v_z \partial f / \partial z = 0, \quad 0 < z < d, \quad (5.1)$$

$$c_0 \partial v_0 / \partial x = -\gamma_1 v_0 + I_{s2}(x), \quad z = 0. \quad (5.2)$$

The boundary condition for the transport Eq. (5.1) on the  $z = 0$  surface can be written down using Eq. (2.13) and the results of Ref. 1, which gives

$$|v_z| f^+(v) = \int_{v_z < 0} d v' |v_z'| f^-(v') [W_0(v' \rightarrow v; T) + \Delta W(v' \rightarrow v; v_0(x))]. \quad (5.3)$$

Here,  $W_0$  is the probability of the scattering of a molecule on the equilibrium surface (see Refs. 15, 16, and 1) and  $\Delta W$  is the part of the scattering probability associated with a non-equilibrium state of a solid during propagation of a high-frequency surface acoustic wave. Reflection of molecules from the  $z = d$  surface will, for the sake of simplicity, be regarded as diffuse and the boundary conditions will be written in the form

$$f_{z=d}^- = f_0^-. \quad (5.4)$$

Here,  $f_0^-$  is a local-equilibrium distribution for molecules with a velocity  $v_z < 0$  and with a coordinate-dependent density  $n_M^-$ .

We shall consider the case when  $d \ll \lambda_0$  and  $\lambda_0 = c_0(\gamma_1 + \gamma_2)^{-1}$  is the attenuation (absorption) length of an acoustic wave in a solid, which is of the order of 1 cm for  $\omega_0 \sim 10^{10}$  Hz at  $T \sim 300$  K (Ref. 17). Then, in the first approximation in respect of the small parameter  $d/\lambda_0$  we can assume that in each transverse cross section of the channel there is a local distribution of molecules with macroscopic parameters which vary slowly along the  $x$  axis. This makes it possible to solve the transport Eq. (5.1) together with the boundary conditions (5.3) and (5.4) for a fixed value of  $v_0$  and then seek the dependences of the macroscopic parameters of the gas on  $x$  using the transport Eq. (5.2) and the condition of conservation of the total gas flux along a channel under steady-state conditions.

We shall initially assume that this channel connects two large volumes of a gas at a temperature  $T$  and a density  $\bar{n}_M$ , and that the channel length  $L_c$  satisfies the condition  $d \ll L_c \ll \lambda_0$ . Then, we can ignore the attenuation of sound along the channel and the problem reduces to the solution of the system of equations (5.1), (5.3), and (5.4), where  $v_0$  has a constant value governed by the power of the source of sound. We shall also bear in mind that the change in the tangential component of the momentum of a molecule due to the absorption of an acoustic phonon does not exceed the quasimomentum of that phonon  $\sim \hbar\omega_0/c_0$ , which is much less than the average momentum of a molecule  $\sim (mT)^{1/2}$ .

Then, the distribution function  $f$  can be sought in the following form:

$$f^\pm(\mathbf{v}, z) = f_0^\pm(v) [1 + \varphi^\pm(\mathbf{v}, z)], \quad |\varphi^\pm| \ll 1. \quad (5.5)$$

If in the solution of Eqs. (5.1), (5.3), and (5.4) we confine ourselves to the approximation linear in  $\varphi$  and calculate the total flux  $J_1$  of molecules per unit width of the channel, we obtain

$$J_1 = d \int_{v_x > 0} dv \frac{v_x}{v^2} \int_{v_z, v_z' < 0} dv' f_0^-(v') \Delta W(\mathbf{v}' \rightarrow \mathbf{v}; v_0). \quad (5.6)$$

In the determination of  $\Delta W$  we shall use in Eq. (5.6), as in Sec. 4, the one-phonon approximation for  $W_D$  (Refs. 19 and 14). Then, employing Eqs. (4.1) and (4.2), we can write down the nonequilibrium part  $\Delta W$  of the kernel of the boundary condition given by Eqs. (2.13) and (5.3) in the form (see also Ref. 1):

$$\begin{aligned} \Delta W(\mathbf{v}' \rightarrow \mathbf{v}; \bar{v}_0) \\ = [1 + P_\Sigma(\mathbf{v}')/4]^{-2} [P^+(\mathbf{v}' \rightarrow \mathbf{v}; \mathbf{K}_0 \xi_0) + P^-(\mathbf{v}' \rightarrow \mathbf{v}; \mathbf{K}_0 \xi_0)]. \end{aligned} \quad (5.7)$$

Here,  $\bar{v} \partial = \bar{R} / \hbar \omega_0$ , where  $\bar{R}$  is the density of the acoustic energy near the source of surface acoustic waves. If we follow the previous procedure and calculate  $P^\pm$  and  $P_\Sigma$  using the model of a continuum with a free boundary of a solid and the molecule-surface interaction potential given by Eq. (4.3) and if we substitute Eq. (5.7) into Eq. (5.6), we readily find that in the first approximation with respect to a small parameter  $\hbar \omega_0 / T$  the expression for the average velocity  $\langle v_x \rangle$  of the flow of a gas in such a channel is

$$\langle v_x \rangle = \frac{J_1}{d \bar{n}_M} = R \Phi \frac{\omega_0}{\rho_s c_0^2} \left( \frac{T}{\pi \hbar \omega_0} \right)^{1/2}. \quad (5.8)$$

For the values of  $\bar{R} \sim 10$  erg/cm<sup>2</sup> (Ref. 17) and  $T \sim 300$  K typical of  $\omega_0 \sim 10^{10}$  Hz the value of  $\langle v_x \rangle$  given by Eq. (5.8) amounts to tens of centimeters per second.

It should be pointed out that the frequency dependence of the gas flow velocity  $\langle v_x \rangle \propto \omega_0^{1/2}$  differs from the frequency dependence of the energy flux  $Q_g \propto \omega_0$  (see Sec. 4) governed by the same microscopic mechanism. This difference is due to the fact that the gas is set in motion because of a correlation between the motion of molecules in the course of scattering along directions normal and tangential to the surface. The tangential motion is due to the simultaneous conservation of the total energy and the tangential momentum (quasimomentum) in the molecule-phonons system and it is responsible for the inequivalence of the various directions of the velocity vectors of molecules reflected in the plane of the surface. On the other hand, such a correlation is unimportant in the determination of the energy flux  $Q_g$  normal to the surface, when obviously it is sufficient to consider the total change in the energy of a molecule and not its redistribution between the various degrees of freedom.

We can also show that Eq. (5.8) is identical with the result obtained in a similar manner by calculation of  $W_D$  of Eq. (2.11) in the first Born approximation using the method of distorted waves. This is due to the fact that the integral of Eq. (5.6) is dominated by the contribution of molecules with sufficiently low normal components of the velocity  $|v_z'| \propto (\hbar \omega_0 / m)^{1/2}$  and the scattering of these simply annihilates

acoustic phonons, whereas creation of phonons is forbidden by the laws of conservation of energy and of the tangential momentum. The special role of the "grazing" molecules, i.e., those incident on the surface at low angles in the drag of the gas by sound is analogous to the corresponding role of grazing electrons<sup>22</sup> in the processes of surface transport in metals. It follows from Eq. (4.5) that in the case of these molecules we have  $P_\Sigma \propto (\hbar \omega_0 / T)^{1/2} \ll 1$  and the above one-phonon approximation is identical with the first Born approximation in the method of distorted waves [see Eqs. (4.1) and (5.7) and also Ref. 14]. We can now understand why the velocity  $\langle v_x \rangle$  of Eq. (5.8) is independent of the mass of the molecules  $m$ . In fact, the velocity acquired by a molecule on absorption of an acoustic phonon is proportional to  $\hbar \omega_0 / mc_0$  and the probability of such a process considered in the first Born approximation used in the method of distorted waves, is proportional to  $m$ .

In the case of a closed channel of arbitrary length (but such that  $d \ll L_c$ ) a macroscopic manifestation of the drag of the gas is the establishment of a certain gas density profile  $n_M(x)$  along the channel. Then, the flux of molecules  $J_1(x)$  governed by Eq. (5.6) with the coordinate-dependent value  $v_0(x)$  is compensated in each section of the channel by an equal and opposite flux  $J_2(x)$  due to the gas density gradient  $\nabla n_M(x)$ . The values of  $J_2(x)$  and  $\nabla n_M(x)$  are related by the transport coefficient  $L$ , which is found by solving the system of equations (5.1), (5.3), and (5.4) subject to  $\Delta W = 0$  allowing for molecular collisions. Using the familiar methods of the kinetic theory of gases,<sup>23,1</sup> we obtain

$$J_2(x) = -L(x) \frac{\nabla n_M(x)}{n_M(x)} = -d^2 \left( \frac{2T}{m} \right)^{1/2} (\alpha_1 \ln Kn + \alpha_2 + \alpha_3). \quad (5.9)$$

Here,  $\alpha_1$ ,  $\alpha_2$ , and  $\alpha_3$  are constants of the order of unity and the values of  $\alpha_1$  and  $\alpha_2$  depend on the intermolecular interaction law; the value of  $\alpha_3$  is governed by the probability  $W_0$ . Equating  $J_1(x)$  of Eq. (5.8) to  $J_2(x)$  of Eq. (5.9), we obtain

$$\begin{aligned} \frac{dn_M(x)}{dx} = n_M(x) v_0(x) \Phi \frac{\omega_0}{\rho_s d c_0^2} \left( \frac{\hbar \omega_0 m}{2\pi} \right)^{1/2} \\ \times (\alpha_1 \ln Kn + \alpha_2 + \alpha_3)^{-1}. \end{aligned} \quad (5.10)$$

When Eq. (5.10) is solved together with the transport equation (5.2), we can determine the dependence of  $n_M$  and  $R$  on the coordinate  $x$  along the channel. Substituting in Eq. (5.10) the values  $\omega_0 \sim 10^{10}$  Hz,  $v_0 = R / \hbar \omega_0 \sim 10^{18}$ ,  $n_M \sim 10^{19}$  cm<sup>-3</sup>, and  $d \sim 10^{-2}$  cm (under these conditions we have  $d \ll \lambda_0 \sim 1$  cm), we can estimate the upper limit to the order of magnitude of the relative change in the density of a gas along a channel  $(\nabla n_M / n_M) \lambda_0 \sim 10^{-2}$ . Then, the solution of the system of equations (5.2) and (5.10) can be obtained by the method of successive approximations in respect of a small deviation of the gas density from the equilibrium value  $\bar{n}_M$ . In the zeroth approximation we can ignore the change in the gas density in Eq. (5.2); the result is

$$v_0(x) = \frac{\bar{R}}{\hbar \omega_0} \exp \left[ -\frac{x}{c_0} (\gamma_1 + \gamma_2) \right], \quad x > 0. \quad (5.11)$$

(It is assumed that the source of surface acoustic waves is located near the end of the channel at a point  $x = 0$ .) Substi-



tuting Eq. (5.11) into Eq. (5.10), we find in particular that in the case of a long ( $\lambda_0 \ll L_c$ ) channel the zeroth approximation in respect of the parameter  $\lambda_0/L_c \ll 1$  gives the following solution

$$n_M(x) = \tilde{n}_M \left\{ 1 - \frac{\tilde{R}\Phi\omega_0}{\rho_1 dc_0} \left( \frac{m}{2\pi\hbar\omega_0} \right)^{1/2} (\gamma_1 + \gamma_2)^{-1} \times (\alpha_1 \ln Kn + \alpha_2 + \alpha_3)^{-1} \exp \left[ -\frac{x}{c_0} (\gamma_1 + \gamma_2) \right] \right\}. \quad (5.12)$$

It follows from Eq. (5.12) that the pressure drop at the ends of the channel is independent of its length and for the above parameters of the system it reaches  $\Delta p/p \sim 10^{-2} - 10^{-3}$  which is easy to determine experimentally.

It should be stressed that the appearance of flow of a gas in a channel under the influence of surface acoustic waves is a fundamental feature of the investigated mechanism of the absorption of surface acoustic phonons by molecules. We can readily show that this effect is absent if we consider the mechanism of the interaction of a molecule with a local-equilibrium macroscopically vibrating surface when surface acoustic waves are propagating. We can therefore predict the temperature dependence of the gas flow velocity  $\langle v_x \rangle$  in the gigahertz frequency range. When the value of  $T$  is low so that the condition  $\omega_0\tau_p \gg 1$  is satisfied, the velocity  $\langle v_x \rangle$  increases in accordance with the law  $\langle v_x \rangle \propto T^{1/2}$  deduced from Eq. (5.8) at sufficiently high values of  $T$ , when  $\omega_0\tau_p \ll 1$ , and the velocity is  $\langle v_x \rangle = 0$ . In the case of intermediate frequencies the velocity  $\langle v_x \rangle$  is a decreasing function of temperature (Fig. 3). The experimental confirmation of this dependence would represent an additional (see Sec. 4) confirmation of the phonon mechanism of the absorption of sound by a gas.

We shall conclude by noting that, as demonstrated above, the transport of the mass (drag effect) and energy in the course of absorption of acoustic phonons by molecules is due to a nonequilibrium state of the surface related to the excess population of an acoustic vibrational mode of a solid. Then, the transport of mass and energy in a gas and the flow of energy (acoustic phonons) in a solid are correlated. Therefore, the transport phenomena discussed above belong to the class of interphase nonlocal transport effects.<sup>1</sup> However, in respect of the nature of an external perturbation acting on a system (which is the excitation of sound) they differ fundamentally from the phenomena in a thermal nonequilibrium system<sup>24</sup> which are discussed in Ref. 1. For example, in the case of propagation of high-frequency sound the interphase nonlocal transport effects appear in systems of normal geometric scale. For comparison, we must mention that in the case of the thermal interaction in a gassolid system the interphase effects are important only in the case of very thin ( $\sim 100 \text{ \AA}$  in diameter) channels.<sup>1</sup> A specific

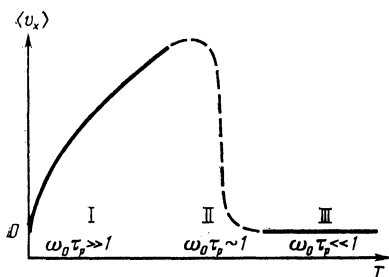


FIG. 3.

feature of the interphase phenomena in the course of propagation of surface acoustic waves is the fact that an external perturbation disturbs from equilibrium only the quasitwo-dimensional subsystem of surface phonons. The macroscopic flow of a gas which is then generated is entirely due to the interphase interaction between the two-dimensional (surface phonons) and three-dimensional (gas) subsystems. On the other hand, in the case of the thermal interaction with a system the transport processes in a gas are governed<sup>1</sup> by a superposition of interphase effects which are due to a nonequilibrium of the subsystems of surface and volume phonons, and also due to the single-phase effect associated with the nonequilibrium of the gas itself.

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<sup>1</sup> It is known<sup>14</sup> that the first Born approximation in the method of distorted waves gives rise to a nonnormalized probability  $W_D$  and gives a reasonable result only for molecules of small masses (for example He), when the total probability of nonspecular reflection is considerably less than unity.

<sup>2</sup> In the case of an arbitrary law governing the reflection of molecules from the surface, we can show that Eq. (4.7) contains an additional factor representing accommodation of the energy and (tangential) momentum of a molecule on the surface and generally dependent on  $m$ . The accommodation coefficients usually rise on increase in  $m$  (Refs. 14 and 21). Therefore, an allowance for the deviation of the law of reflection of molecules from the diffuse mechanism only results in a deterioration of the agreement with the experimental results at the lowest of the investigated<sup>2</sup> frequencies (see Fig. 1) and cannot account for the deviation which increases on increase in  $\omega_0$ , from the dependence  $\gamma_2 \propto \omega_0 m^{1/2}$ .

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