Surface phenomena on the interface of solid and superfluid ^{3}He–^{4}He solutions

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(Submitted 24 January 1986)
Zh. Eksp. Teor. Fiz. **91**, 826–836 (September 1986)

A complete system of hydrodynamic boundary conditions on the interface of solid and superfluid ^{3}He–^{4}He solutions is formulated. This system takes into account the finite ^{3}He concentration in both the liquid and the solid phases. The spectrum and damping of the melting-crystallization waves are analyzed. The Čerenkov contribution of the thermal melting-crystallization waves and of the Rayleigh waves to the impurity part of the Kapitza thermal resistance is calculated. The inverse effect associated with the possibility of resonant excitation by an internal impurity second sound wave of surface Rayleigh and crystallization waves is also considered.

The interface between solid and superfluid ^{4}He is an unparalleled object for the investigation of dynamic surface phenomena. Interest in this interface is due above all to its anomalously high mobility, predicted by Andreev and Parshin.[1] This prediction was later experimentally confirmed in a number of studies. Keshishev, Parshin, and Babkin[2] investigated the spectrum and damping of the melting-crystallization waves. Castaign and Nozieres,[3] Castaign, Balibar, and Laroche,[4] and Grigor'ev, Dyumin, and Svatko[5] studied the conversion of sound passing through such an interface. Maris and Huber,[6] Puech *et al.*,[7] and Wolf *et al.*[8] measured the Kaptiza thermal resistance at the interface between helium and He II. Finally, Graf, Bowley, and Maris[9,10] recently investigated the influence of a small amount of ^{3}He impurity on the mobility of the interface and on its thermal resistance. It was established in these experiments, in particular, that at low temperature the interface of crystals with dimensions L larger than the capillary constant $a_0 \approx 0.14$ cm and with ^{3}He concentration up to 10^{-3} remains highly mobile. This may seem to contradict the results of Landau *et al.*[11] who observed visually an increase of the relative flat surface fraction on small ($L \sim \alpha_0$) crystals when a low-concentration ($\sim 10^{-4}$) impurity is added. Qualitative arguments by Castaing *et al.*,[12] however, attribute this phenomenon to the fact that adsorption of the ^{3}He impurity on the surface lowers the surface rigidity $\tilde{\alpha} = \alpha + \partial^2\alpha/\partial\varphi^2$ and thereby also the sizes of the rounded sections. For large crystals ($L \gg a_0$), however, the role of surface energy in the establishment of the equilibrium shape is small compared with that of gravity, so that when a low-concentration impurity is added an appreciable fraction of the boundary remains highly mobile.[10] In Refs. 9 and 10 the authors have also proposed for the interaction of ^{3}He with the interface a theoretical model involving a macroscopically extended wave function of the impurity quasiparticle, and using the assumption that all the ^{3}He is drawn to the interface that is set oscillating by the thermal phonons. This assumption is in fact equivalent to neglect of the equilibrium "freezing-in" (penetration) of the impurity in the solid phase. It can be deduced from the experiments of Anufriev, Lopatik, and Sebedash[13] and also from the stratification curves for the solid and liquid solutions at pressure $P \approx 25$ atm,[14,15] that the equilibrium concentation of the impurity in the solid solution is indeed negligible compared with the concentation in the liquid at temperatures $T > 0.3$ K. At $T \sim 0.4$–0.5 K, however, the equilibrium impurity densities in the solid and liquid phases become comparable in order of magnitude.

We derive here a complete system of hydrodynamic boundary conditions on the interface of solid and superfluid ^{3}He–^{4}He solutions in the phonon temperature region $T \lesssim 0.5$ K. These conditions take into account the finite ^{3}He concentration in both the liquid and solid phases. The system contains two more equations than in the case of the interface of pure solid and liquid ^{4}He (Refs. 2, 16–19), viz., conservation of the number of impurity particles and thermodynamic equilibrium in the impurity component. We emphasize that the system is self-consistent (in the sense that the equations and variables are equal in number) only if account is taken of the dissipative mechanism of heat and impurity transport. It extends also the known boundary conditions for weak-non-equilibrium processes on the interface of classical solid and liquid solutions[20] to include the quantum case of low temperatures and a superfluid liquid phase. The resulting boundary conditions are used to analyze the influence of the impurity on the damping of the melting-crystallization waves.

We propose also a mechanism different from that of Refs. 9 and 10 for the heat transfer from a ^{4}He crystal to a gas of ^{3}He quasiparticle. This mechanism consists of energy transfer from a phonon in the solid to a second-sound quantum in the liquid solution. In a sufficiently large temperature interval and at relatively low densities, second sound constitutes a wave of coupled temperature and concentration oscillations of an ideal nondegenerate ^{3}He-quasiparticle gas (second sound is sound in an impuriton gas).[21] The impurity contribution to the Kapitza thermal conductivity σ_{S3} of this interface is threfore proportional to the small coefficient of conversion of first-sound energy in the solid into second-

0038-5646/86/090487-06$04.00

sound energy in the liquid solution. Calculation shows that this coefficient is independent of frequency and its order of magnitude is such that it can be extrapolated to the high-frequency region of the ballistic regime of the impurity oscillations. This mechanism is in fact similar to Khalatnikov's well-known[22] mechanism of energy exchange between the phonons of a solid and the He II phonons.

We point out in the second part of the paper that, in view of the ordering of the phase velocities of the first and second sound, the second-sound wave can resonantly excite in the system Rayleigh waves and melting-crystallization waves. This leads accordingly to the appearance of two different angles beyond the critical value, near which the energy reflection coefficient differs noticeably from unity.

Lastly, we obtain the contribution of the Čerenkov emission of second-sound quanta by the thermal crystallization oscillations of the boundary and by the Rayleigh waves in the impurity component of the Kaptiza thermal conductivity σ_{S3}. Since the thermal crystallization waves have near $T \gtrsim 0.1$ K a linear dispersion due to the effective mass,[23,24] this contribution has the same dependence on temperature and concentration, and the same order of magnitude, as the contribution, investigated in Refs. 9 and 10, from the direct energy transfer from the bulk thermal phonon to the impurity subsystem.

BOUNDARY CONDITIONS ON THE INTERFACE OF SOLID AND SUPERFLUID ^{3}He–^{4}He SOLUTIONS

To derive the boundary conditions, we use, in the linear approximation, the expressions for the bulk fluxes of matter $\mathbf{j}^{(1,2)}$, impurity $\mathbf{j}_3^{(1,2)}$, momentum $\Pi_{ik}^{(1,2)}$, and entropy $\mathbf{f}^{(1,2)}$. The superscripts 1 and 2 refer to the solid and liquid phases, respectively. The fluxes in the liquid phase are of the form[22]

$$\mathbf{j}^{(2)}=\rho_s\mathbf{v}_s+\rho_n\mathbf{v}_n, \quad \mathbf{j}_3{}^{(2)}=\rho_2 c_2\mathbf{v}_n,$$
$$\Pi_{ik}^{(2)}=P^{(2)}\delta_{ik}, \qquad \mathbf{f}^{(2)}=\rho_2\sigma_2\mathbf{v}_n-\varkappa_2\nabla T_2/T_2, \tag{1}$$

where ρ, ρ_s, ρ_n, $\mathbf{v}_n$, $\mathbf{v}_s$, P, σ, and T have the meaning standard in superfluid hydrodynamics, c is the molar concentration of the impurity, and $\varkappa_2$ is the thermal conductivity of the solution.

It follows from theroretical and experimental data that in the temperature and concentration region under consideration, $T \sim 0.1$–0.5 K, $c_2 \sim 10^{-3}$–10^{-4}, the influence of diffusion, including heat and mass diffusion, is negligible in the liquid phase compared with the thermal conductivity, and will hereafter be neglected. Recall that the expression for the normal density and entropy of a weak nondegenerate superfluid solution are of the form

$$\rho_n=\rho_n{}^{ph}+\rho_2 c_2 m^*/m_4 \tag{2}$$

(if both the impurity and the phonon subsystems oscillate in the hydrodynamic regime),

$$\sigma_2=\sigma_2{}^{ph}-\frac{c_2}{m_4}\ln c_2+\frac{c_2}{m_4}\ln\left[\frac{2m_4}{\rho_2}\left(\frac{m^*T}{2\pi\hbar^2}\right)^{3/2}\right], \tag{3}$$

where $\rho_n^{\rm ph}$, $\sigma_2^{\rm ph}$ are the phonon contributions to the normal density and to the entropy, m_3 is the mass of the ^{4}He atom, and m^* is the effective mass of the ^{3}He atom in the solution.

The expressions (1) for the fluxes are in accord with the law $\dot{E}_2+\operatorname{div}\mathbf{Q}_2=0$ of energy conservation in the liquid. In the limit of low concentations we have

$$E_2=-P_2+\rho_2 T_2\sigma_2+\mu_4^{(2)}\rho^{(2)}+\mu_3^{(2)}\rho_2 c_2,$$
$$\mathbf{Q}_2=\mu_4^{(2)}\mathbf{j}_2+\mu_3^{(2)}\mathbf{j}_3^{(2)}+T_2\mathbf{f}_2, \tag{4}$$

where $\mu_4^{(2)}$ is the chemical potential of the solvent per unit mass of pure ^{4}He and $\mu_3^{(2)}$ is the chemical potential of the solute. For a weakly nondegenerate solution we have

$$\mu_4^{(2)}(P,T,c_2)=\mu_4^{(2)}(P,T)-(T/m_4)c_2,$$
$$\mu_3{}^{(2)}=\Psi^{(2)}(P,T=0)-\left(\frac{T}{m_4}\right)\ln\left[\frac{2m_4}{\rho_2}\left(\frac{m^*T}{2\pi\hbar^2}\right)^{3/2}\right]+\left(\frac{T}{m_4}\right)\ln c_2. \tag{5}$$

We neglect in (5) the small thermal expansion of the solution. In Eq. (5), $\mu_4^{(2)}(P,T)$ is the chemical potential of pure ^{4}He and $\Psi^{(2)}$ is the binding energy of one impurity atom in liquid ^{4}He.

Note that the hydrodynamic description is valid in the liquid phase for frequencies lower than all the characteristic reciprocal relaxation times in the system. In the temperature and concentration region under consideration we have the following frequency ordering

$$1/\tau_{ph\,ph}^{(2)} \ll 1/\tau_{phi}^{(2)} \ll 1/\tau_{ii}^{(2)}$$

for the phonon-phonon, phonon-impurity, and impurity-impurity relaxation, respectively. At $T \lesssim 0.5$ K, however, we have $c_2 \sim 10^{-4}$–10^{-3}, i.e., hydrodynamics is valid up to quite high frequencies, $\omega \sim 1/\tau_{ii}^{(2)} \sim 10^7$–$10^8$ s^{-1}, in that region where the impurities and their scattering by one another are of principal significance for the thermodynamics and formation of second-sound waves.

Let us dwell in somewhat greater detail on the expressions for the bulk fluxes in the solid phase. The possibility of a complete hydrodynamic description, particularly of introducing a single temperature for the impurity and phonon subsystems, implies satisfaction of the condition $\omega\tau_{i\rm ph}^{(1)}<1$ (the impurity-thermalization condition). We emphasize that at the temperatures and concentrations considered, the following relation holds between the respective relaxation frequencies[25–27]

$$1/\tau_{iph}^{(1)} \ll 1/\tau_{phi}^{(1)} \lessgtr 1/\tau_{phph}^{(1)U} \lessgtr 1/\tau_{ph\,ph}^{(1)N} \lesssim 1/\tau_{ii}^{(1)}$$

for an impurity on a phonon, a phonon on an impurity, a phonon on a phonon with umklapp, and normal scattering of a phonon by a phonon on account of anharmonicity and of an impurity by an impurity due to band motion. At $T \sim 0.5$ K we have $1/\tau_{i\rm ph}^{(1)} \sim 10^2$–$10^3$ s^{-1} and $1/\tau_{\rm phph}^{U} \sim 1/\tau_{\rm phph}^{N} \sim 10^6$ s^{-1}. When all these relations between the frequency and the relaxation times are satisfied, the bulk fluxes in the solid phase take the form

$$\mathbf{j}_1=\rho_1\dot{\mathbf{u}},\quad \mathbf{j}_3^{(1)}=\rho_1 c_1\dot{\mathbf{u}}-\rho_1 D_1\nabla c_1,$$
$$\Pi_{ik}^{(1)}=P_1\delta_{ik}-\sigma_{ik}^{*},\tag{6}$$
$$\mathbf{f}^{(1)}=\rho_1\sigma_1\dot{\mathbf{u}}-\varkappa_1(\nabla T_1/T_1)-(\partial\sigma_1/\partial c_1)\rho_1 D_1\nabla c_1.$$

Expressions (6) for the fluxes are in accord with the law of energy conservation in a solid:

$$\dot{E}_1+\operatorname{div}\mathbf{Q}_1=0,$$
$$E_1=-P_1+\rho_1\sigma_1 T_1+\mu_4^{(1)}\rho^{(1)}+\mu_3^{(1)}\rho^{(1)}c^{(1)}+\sigma_{ik}^{*}u_{ik}^{*}/2,\tag{7}$$
$$\mathbf{Q}_1=\mu_4^{(1)}\mathbf{j}^{(1)}+\mu_3^{(1)}\mathbf{j}_3^{(1)}+T_1\mathbf{f}_1+\boldsymbol{\sigma}_k^{*}\dot{u}_k,$$

where $\sigma_{ik}^{*}=\sigma_{ik}-1/3\sigma_{ll}\delta_{ik}/3$ is the deviator of the stress tensor, $P_1=P_0-1/3\sigma_{ll}/3$, P_0 is the equilibrium pressure on the melting curve, and D_1 and $\varkappa_1$ are the diffusion and heat-conduction coefficients. We point out that in our range of temperatures and concentrations the coefficient D_1 is determined mainly by impurity-impurity collisions for itinerant motion (self-diffusion). Introduction of this coefficient in our equation is justified because the impurity bandwidth is $\Delta\ll T$, meaning high probability of impurity scattering with quasimomentum transfer to the crystal lattice (umklapp).

In the limit of low concentrations we have in (6) and (7)

$$\sigma_1=\sigma_1^{ph}-(c_1/m_4)\ln c_1,$$
$$\mu_4^{(1)}(P,\ T,\ c_1)=\mu_4^{(1)}(P,\ T)-(T/m_4)c_1,\tag{8}$$
$$\mu_3^{(1)}=\Psi^{(1)}(P,\ T=0)+(T/m_4)\ln c_1.$$

We have neglected here the small contribution to the thermodynamics from the slow itinerant impurity [the effective itinerant velocity is $v_b\sim\Delta d/\hbar\ll(T/m^{*})^{1/2}$, where d is the distance between the atoms]. The last circumstance leads to a difference between the expressions for μ_3 in the solid and superfluid solutions in Eqs. (5) and (8).

To derive the boundry conditions, we use the conservation of the total energy, with the surface energy taken in the form[28]

$$\dot{\mathscr{E}}+\nabla_\beta\Theta_\beta-(Q_{1z}-Q_{2z})+(E_1-E_2)V_{\mathrm{rp}}=0,\tag{9}$$

where z is the direction of the normal to the unperturbed interface (the crystal occupies the half-space $z<0$, $\mathbf{Q}_1$, $\mathbf{Q}_2$, E_1, and E_2 are defined by expressions (1), (4), (6), and (7). $\mathscr{E}$ and Θ_β are the surface-energy density and flux, and $\beta=(x,y)$.

Since we shall not be interested in effects due to adsorption of the impurity and to its diffusion over the surface, a detailed definition of the surface (using the condition that either the total surface or its impurity part vanish) is immaterial. In Eq. (9) we have

$$\mathscr{E}=\mathscr{E}_0(T)+{}^1/_2M_{eff}\dot{\xi}^2+{}^1/_2\tilde{\alpha}_{\mu\nu}\xi_\mu\xi_\nu,$$

where M_{eff} is the effective surface mass connected, for example, with the restructuring of the short-range order on the crystal-liquid interface,[23,24] $\tilde{\alpha}_{\mu\nu}=\alpha\delta_{\mu\nu}+\partial^2\alpha/\partial\varphi_\mu\varphi_\nu$ is the surface rigidity tensor, $\Theta_\beta=-\tilde{\alpha}_{\mu\beta}\xi_\mu\xi$ (Ref. 28), $V_{\mathrm{int}}=\dot{u}_z+\dot{\xi}$ ($\mathbf{u}$ is the elastic-displacement vector), and ξ is the displacement of the atom-rough interface in addition to the elastic displacement.

From the condition that the total-energy consevation law agree with the equations for mass conservation

$$j_z^{(1)}-\rho_1 V_{\mathrm{int}}=j_z^{(2)}-\rho_2 V_{\mathrm{int}},\tag{10}$$

and impurity conservation

$$j_{3z}^{(1)}-\rho_1 c_1 V_{\mathrm{int}}=j_{3z}^{(2)}-\rho_2 c_2 V_{\mathrm{int}},\tag{11}$$
$$\Pi_{zz}^{(1)}=\Pi_{zz}^{(2)},\qquad \Pi_{z\beta}^{(1)}=0\tag{12}$$

and from the law of increase of surface entropy

$$f_z^{(1)}-\rho_1\sigma_1 V_{\mathrm{int}}-f_z^{(2)}+\rho_2\sigma_2 V_{\mathrm{int}}=R_{\mathrm{sur}}/T_0\tag{13}$$

we obtain the form of the surface dissipative function R_{sur}:

$$R_{\mathrm{sur}}=[\mu_4^{(1)}-\mu_4^{(2)}+(M_{eff}\ddot{\xi}-\tilde{\alpha}_{\gamma\beta}\nabla_\gamma\nabla_\beta\xi)/\rho_1](j_z^{(1)}-\rho_1 V_{\mathrm{int}})$$
$$+(\mu_3^{(1)}-\mu_3^{(2)})(j_{3z}^{(1)}-\rho_1 c_1 V_{\mathrm{int}})|+(T_1-T_2)(f_z^{(1)}-\rho^{(1)}\sigma^{(1)}V_{\mathrm{int}}).\tag{14}$$

We have neglected in (10)–(14) the surface fluxes of mass, impurity mass, momentum, and entropy, and have set the surface temperature equal to those in the interior. In Eq. (13), T_0 is the equilibrium temperature on the melting curve.

From the requirement that the surface dissipative function be positive-definite we obtain the following generalization of the condition for the thermodynamic phase equilibrium:

$$[\mu_4^{(1)}-\mu_4^{(2)}+(M_{eff}\ddot{\xi}-\tilde{\alpha}_{\gamma\beta}\nabla_\gamma\nabla_\beta\xi)/\rho_1]=(j_z^{(1)}-\rho_1 V_{\mathrm{int}})/K_4,$$
$$(\mu_3^{(1)}-\mu_3^{(2)})=(j_{3z}^{(1)}-\rho_1 c_1 V_{\mathrm{int}})/K_3,$$
$$(T_1-T_2)=R_{\mathrm{K}}T_0(f_z^{(1)}-\rho_1\sigma_1 V_{\mathrm{int}}).\tag{15}$$

We disregard here for simplicity the off-diagonal elements of the Onsager matrix of the kinetic surface coefficients. In the phenomena under consideration the off-diagonal kinetic-coefficient matrix elements (which are smaller than the diagonal ones, since the surface dissipative function is positive-definite) only renormalizes the effects connected with the diagonal elements, and does not lead to qualitatively new results.

In (15), R_K is the Kapitza thermal resistance (it is determined by energy transfer from the interface to the quasiparticle gas). In the case of a superfluid solution, the total thermal conductivity σ_S of the interface is a sum of two contributions, σ_{SL} from the phonons and σ_{S3} from the impurities. The phonon contribution $\sigma_{SL}\propto T^5$ is determined at low temperatures ($T\lesssim 0.2$ K) by capillary effects, viz., the surface rigidity $\tilde{\alpha}$ and the effective surface mass M_{eff}. It was investigated in Refs. 6 and 24. The impurity contribution to the heat conduction of the interface will be discussed below.

In Eq. (15), $1/K_4$ is the reciprocal crystal-growth coefficient (relative to the solvent). In quantum crystals it is determined mainly by the momentum transfer from the quasiparticle gas to the interface.[1,17,18] If the drift velocities of the impurity and of the phonons do not coincide with the interface velocity (as is the case when the impurity has a finite ability to freeze into the solid phase), the $1/K_4$, just like σ_S, is a sum of two contributions—from the impurities and from the phonons. The phonon contribution $1/K_4^{(\mathrm{ph})} \propto \rho_n^{\mathrm{ph}} c_{\mathrm{liq}}/\rho^2 \propto T^4$ (where c_{liq} is the speed of sound in the liquid) was investigated experimentally in Ref. 2. The impurity contribution

$$1/K_4^{(i)} \propto \rho_{ni}(T/m^*)^{1/2}/\rho^2 \propto c^2 T^{1/2}$$

was estimated in Ref. 12. Lastly, $1/K_3$ has the meaning of the coefficient of crystal growth relative to the solute. A similar coefficient was introduced for the growth of a classical crystal, e.g., in Ref. 16. A possible estimate of its value for the interface considered will be given below. When viscosity of the liquid is taken into account, one more condition must be met, equality of the tangential components of the velocity of the normal excitations in the liquid to the lattice velocity $\dot{u}_\beta = v_{n\beta}$. In addition, Eqs. (12) and (14) acquire terms connected with the viscous-stress tensors. For the phenomena considered below, however, viscosity is insignificant and will be neglected.

We discuss now the system (10)–(13) and (15) (consisting of eight equations) to determine whether the number of boundary conditions equals the number of independent modes. These are first and second sounds, and the purely dissipative heat-conduction mode having at $c_2 \sim 10^{-3}$ a spectrum $i\omega_{\mathrm{III}} = 3\varkappa_2 k^2/5\rho_2 C^{(2)}$ ($C^{(2)}$ is the heat capacity per unit mass of the liquid), in which the temperature and the concentation oscillate. If allowance were made for diffusion, the spectrum of this mode would be determined by the effective thermal conductivity $\varkappa_{\mathrm{eff}}$ (see Ref. 22). There is no such mode in pure He II, and the heat conduction influences only the second-sound damping $\mathrm{Im}\,\omega_{\mathrm{II}} = 1/5\varkappa_2 k^2/\rho_2 C^{(2)}$. The presence of an independent heat-conduction mode in the solution is very clearly due to the appearance of an independent variable—the concentration ($\omega_{\mathrm{III}} \to 0$ as $c_2 \to 0$ and at $D_2 = 0$).

Four independent modes exist in the solid. These are two first sounds (longitudinal and transverse), a heat conduction mode $i\omega = \varkappa_1 k^2/\rho_1 C_{\mathrm{ph}}^{(1)}$ (if $\omega\tau_U < 1$) or a second-sound wave in the phonon gas (if $\omega\tau_U > 1$), and last, a diffusion mode $i\omega = D_1 k^2$.

We emphasize that in view of the relation $D_1 \ll \varkappa_1/\rho_1 C^{(1)}$, the heat-conduction and diffusion modes interact very weakly even when thermal diffusion is taken into account. Therefore the stringent impurity-thermalization condition $\omega\tau i_{\mathrm{ph}}^{(1)} < 1$ is, generally speaking, not mandatory for the hydrodynamic scheme being considered. Hydrodynamic treatment of diffusion and heat conduction in a solid is thus actually valid up to frequencies $\omega < 1/\tau_{\mathrm{phph}}^{N(1)} \lesssim 1/\tau_{ii}^{(1)}$, i.e., to $\omega \sim 10^6\ \mathrm{s}^{-1}$. The eighth independent variable is the additional-recrystallization displacement ξ of the interface. The number of independent variables is therefore equal to the number of the boundary conditions.

DAMPING OF MELTING-CRYSTALLIZATION WAVES

The specific property of the interphase boundary in question is that three surface modes exist on it: melting-crystallization waves (in which ξ and $\mathbf{j}^{(2)}$ oscillate), Rayleigh waves (in which $\mathbf{u}$ and ξ oscillate), and surface second sound, whose velocity is close to that of the bulk second sound in He II (in which coupled oscillations of the temperature and concentration take place) (see Ref. 29).

Using the derived boundary conditions, let us examine how a low concentration of ^{3}He impurity influences the damping of the crystallization waves.

The spectrum of the melting-crystallization waves is of the form

$$\omega^2 = \frac{\tilde{\alpha}_{xx} k^3 \rho/(\Delta\rho)^2}{1+(\rho\rho_n/\Delta\rho)^2} - i\omega k\rho\left(\frac{\rho}{\Delta\rho}\right)^2 \left[\frac{1}{K_4} + \frac{c_1^2}{K_3} + R_{\mathrm{K}} T \sigma_1^2 + \frac{T(\sigma_1 - \sigma_2 c_1/c_2)}{(\varkappa_1+\varkappa_2)k}\left(\sigma_{1\mathrm{ph}} + \frac{c_1}{m_4}\ln\frac{c_2}{c_1}\right)\right], \tag{16}$$

where k is the wave number of the surface wave.

At $T \sim 0.4$–0.5 K the impurity, in both the liquid and solid phase, makes a larger contribution to the thermodynamics than the phonons. The imaginary part of the wave spectrum can therefore be written in this case in the form

$$\mathrm{Im}\,\omega = \frac{\rho k}{2}\left(\frac{\rho}{\Delta\rho}\right)^2 \left\{\frac{1}{K_4} + \frac{c_1^2}{K_3} + R_{\mathrm{K}} T \sigma_1^2 + \frac{T c_2^2}{m_4^2 k(\varkappa_1+\varkappa_2)}\left(\frac{c_1}{c_2}\right)^2 \ln^2\frac{c_1}{c_2}\right\}. \tag{17}$$

The first three terms in the right-hand side of (17) are responsible for the surface dissipation, and the fourth for the bulk dissipation. Estimates show that for

$$k l_U < (u_{\mathrm{II}}/C_{\mathrm{liq}})(\sigma_i/\sigma_{\mathrm{ph}}) c_1/c_2 \tag{18}$$

(where C_{liq} and u_{II} are the velocities of the first and second sounds in the liquid, and σ_i is the impurity part of the entropy) the main contribution to the damping is made by the bulk dissipation. Note that from the damping of the crystallization waves one can determine independently through experiment the impurity distribution coefficient c_1/c_2, which is a function only of temperature and pressure in weak solutions.

We point out that if the impurity concentration is decreased in each of the phases, the heat-conduction contribution to the imaginary part of (16) at a fixed temperature is determined by the phonons and differs from zero. If, however, the inequality (18) is reversed, the damping is determined mainly by surface dissipation. Note that for $T \sim 0.4$–0.5 K and starting with a concentration $c_2 \sim 10^{-3}$ the imaginary part of the spectrum becomes of the same order as the real part (for frequencies $\omega \lesssim 10^5\ \mathrm{s}^{-1}$).

Equation (16) was obtained under the assumption

$$R_{\mathrm{K}} \ll \left(\frac{1}{\varkappa_1 k} \sim \frac{1}{\varkappa_2 k}\right), \qquad R_{\mathrm{K}} \gtrsim \frac{\omega}{k u_{\mathrm{II}}^2 \rho C^{(1)}},$$
$$\frac{1}{K_3}(D_1\omega)^{1/2} \ll \frac{T}{\rho m c_1}, \qquad \frac{c_1^2}{K_3} \lesssim \frac{T c_2^2}{m_4^2 k(\varkappa_1+\varkappa_2)}\left(\frac{c_1}{c_2}\right)^2 \ln^2\frac{c_1}{c_2}. \tag{19}$$

If the first inequality of (19) is violated, there is no flow of entropy through the interface, i.e.,

$$f_{1z} - \rho_1\sigma_1 V_{\mathrm{int}} = f_{2z} - \rho_2\sigma_2 V_{\mathrm{int}} = 0.$$

We emphasize that in the case of pure He II, i.e., for $\omega_{\mathrm{III}} = 0$, the dissipative term in the liquid-phase entropy flux turns out to be small and we arrive at the "stringent" hydrodynamic boundary condition $v_{nz} = V_{\text{int}}$ used in Refs. 17 and 18. When the third and fourth inequalities of (19) are violated, there is no impurity flux through the interface, i.e.,

$$j_{3z}^{(1)} - \rho_1 c_1 V_{\text{int}} = j_{3z}^{(2)} - \rho_2 c_2 V_{\text{int}} = 0,$$

which is also equivalent, for the liquid phase, to the condition $v_{nz} = V_{\text{int}}$. In this case the imaginary part of the crystallization-wave spectrum is given by

$$\operatorname{Im} \omega = \frac{1}{2} k \left(\frac{\rho}{\Delta\rho} \right)^2 \rho \left\{ \frac{1}{K_4} + \frac{Tc_1}{\rho m_4 (D_1 \omega)^{1/2}} \right\},$$

i.e., it is determined at very low frequencies by the diffusion coefficient D_1. Naturally, this contribution vanishes together with the impurity concentration in the solid phase. For the reciprocal of the growth coefficient in the solute we can obtain the estimate

$$\frac{1}{K_3} \sim \frac{T}{\rho m c_1} \frac{l_{ii}^{(1)}}{D_1 \eta} \sim \frac{T}{\rho m c_1} \frac{1}{v_b \eta},$$

where the dimensionless parameter $\eta < 1$ plays the role of the effective coefficient of penetration from the solid into the liquid phase, and becomes much less than unity at temperatures $T < 0.3$ K, when the ability of the impurity to freeze into the solid is low. We emphasize that the transition to the situation in which there are no entropy and impurity fluxes through the interface can be described not only kinetically but thermodynamically. The transition occurs at

$$c_1 \ll c_2, \sigma_{1\text{ph}} \ll (\sigma_2 \approx \sigma_{2i}). \tag{20}$$

Estimates show that both inequalities in (20) hold at the same temperatures $T \sim 0.1$–0.2 K. At these temperatures the solid can be regarded as free of impurities and phonons, i.e., it can be described by the elasticity-theory equations for $T = 0$.

We note in conclusion that at low temperatures (when the impurity concentation c_1 in the solid phase is vanishingly small) the damping of the crystallization waves is determined mainly by the phonon contribution, and the growth coefficient $1/K_4$ is proportional to T^4 and is small. In this case a more important role can be assumed by the corrections (connected with the impurity concentration in the liquid phase) to the real part, and particularly by the effective decrease of the surface rigidity [see Eq. (16)].

ANOMALOUS REFLECTION OF SECOND SOUND FROM THE INTERFACE IN THE CASE OF INCIDENCE AT ANGLES GREATER THAN THE CRITICAL ANGLE

At frequencies that are not too low, there exists in the system considered the following velocity ordering:

$$u_{\text{II}} < v_M < C_R < C_t < C_{\text{liq}} < C_l, \tag{21}$$

where $v_M(\omega)$ and C_R are the phase velocities of the crystallization and Rayleigh waves, and C_t and C_l are the transverse and longitudinal sound velocities in the solid. A critical angle $\sin \theta_{\text{cr}} = u_{\text{II}}/C_t$ exists therefore for the incidence of second sound on the interface. In the absence of dissipation, if the second sound were to be incident at angles greater than critical, total internal reflection would be observed. At the same time, however, Rayleigh surface waves and melting-crystallization waves are present in addition to the bulk waves. If their frequency and wave-vector component tangent to the surface coincide with the corresponding parameters of the bulk second-sound wave incident on the interface [this is permitted by relations (21)], the resonance condition is met. The smaller Rayleigh angle ($\sin \theta_R = u_{\text{II}}/C_R$) is independent of the frequency of the incident wave. The angle of resonant excitation of the crystallization waves is $\sin \theta_M = u_{\text{II}} [\Delta \rho^2 / \tilde{\alpha}_{xx} \rho \omega]^{1/3} = u_{\text{II}}/v_M$. This angle becomes real ($\sin \theta_M < 1$) only at sufficiently high frequencies, $\omega \sim 10^7\ \text{s}^{-1}$ for $T \sim 0.4$ K.

Thus, the presence of a crystallization wave along with a Rayleigh wave in the system leads to the existence of two transcritical angles of anomalous reflection from the considered interface.

We emphasize that resonance can occur in an acoustic experiment only if dissipation is introduced into the system. When account is taken, for example, of surface dissipation, the law of energy conservation in a second-sound wave incident on the interface at a transcritical angle takes the form

$$1 - r_{\text{II}} = R_{\text{sur}} / \rho_n u_{\text{II}} \cos \theta_{\text{II}} \, |v_{n\text{II}}^{\text{inc}}|^2,$$

where $r_{\text{II}} = |v_{n\text{II}}^{\text{ref}}|^2 / |v_{n\text{II}}^{\text{inc}}|^2 < 1$ is the second-sound reflection coefficient, $\theta_{\text{II}} > \theta_{\text{cr}}$ is the incident angle, and $R_{\text{sur}} > 0$ is the surface dissipation function (14). Thus, surface dissipation can decrease noticeably the transmission coefficient. The bulk dissipative mechanisms considered in the preceding section can also renormalize the reflection coefficient for incidence at angles beyond critical.

Transcritical surface-wave resonant-excitation angles exist also in pure He II at higher temperatures, $T \sim 0.8$–1 K. They are caused by the lower velocity of the roton second sound, and hence by the possibility of satisfying the condition $u_{\text{II}} < C_R$ (low-frequency crystallization waves are strongly damped at these temperatures).

Note that Eq. (21) shows that Rayleigh waves cannot be excited on this interface by a first-sound wave in a liquid (in contrast to the case of an He II–metal interface).

IMPURITY CONTRIBUTION TO THE KAPITZA THERMAL RESISTANCE AT LOW TEMPERATURES

Using the complete system of boundary conditions in the low-temperature limit $T \sim 0.1$–0.2 K ($c_1 \ll c_2$, $\sigma_{1\text{ph}} \ll \sigma_2$, $v_{nz} \approx V_{\text{int}}$), we solve the acoustic problem of determining the coefficient of energy transfer by a first-sound wave from a solid to first- and second-sound waves in a liquid. For the simplest case of normal incidence of a longitudinal wave, this coefficient is equal to

$$t_{\text{comp}} = t_{\text{I}} + t_{\text{II}} = 4 \left[\frac{C_{\text{liq}}}{C_l} \varepsilon^2 + \frac{\rho \rho_n u_{\text{II}}}{(\Delta\rho)^2 C_l} \right], \tag{22}$$

where

$$\varepsilon = \frac{\rho M_{eff}}{(\Delta\rho)^2} \frac{\omega}{C_{\text{liq}}}, \quad \rho \approx \rho_1 \approx \rho_2, \quad \frac{\Delta\rho}{\rho} = \frac{\rho_1 - \rho_2}{\rho} \ll 1.$$

Equation (22) contains a sum of two contributions. The first, t_{I}, describes the energy-flux fraction transferred to first sound in the liquid, and t_{II}, the fraction transferred to second sound. In the range of temperatures and concentrations ρ_n which we are considering, u_{II} is determined by the impurity, and the coefficient t_{II} is given by

$$t_{II}=\frac{4\rho}{(\Delta\rho)^2C_l}\left(\frac{5}{3}m^*T\right)^{1/2}\frac{\rho c_2}{m_4}. \qquad (23)$$

Equation (23) does not depend on frequency and does not contain the relaxation times τ. The expression for t_{II} can therefore be used to represent (in order of magnitude, at least) the efficiency with which a thermal phonon $\hbar\omega\sim T$ is transferred to the impurity subsystem. The physical reason is that the Kapitza thermal resistance is determined by the energy transfer from the wall to the He II quasiparticles on the interface itself, so that R_K depends little on the excitation mean free path l (on the relaxation time τ). The path l determines only the distance over which the temperature jump is formed (localized), and not the magnitude of this jump. We thus obtain the following estimate for the impurity contribution to the thermal conductivity σ_{S3} of the interface.

$$\sigma_{s3}\sim C^{(1)}_{(\mathrm{ph})}\overline{C}_t t_{II}\propto(\rho/\Delta\rho)^2c_2T^{7/2},$$

where $C^{(1)}_{\mathrm{ph}}$ is the phonon part of the heat capacity of the solid and $\overline{C}_t$ is the average sound velocity in the crystal. We point out that the resulting temperature and concentration dependences of σ_{S3} agree with the result of the model calculation.[9,10]

[It is curious that the energy fraction transferred to the impurity subsystem by longitudinal sound, obtained in Ref. 10 within the framework of the present quantum-mechanical model (the case $l=\infty$), differs from (23) (the case $l=0$) by no more than 20–25%.]

We point out that t_I of (22) determines the phonon contribution to the thermal conductivity of the interface (σ_{SL}). The latter, by virtue of the proportionality of t_I to ω^2, takes the form $\sigma_{SL}\propto(\rho/\Delta\rho)^4T^5$. The total thermal conductivity $\sigma_S=\sigma_{SL}+\sigma_{S3}$ of the interface is an additive quantity, a fact reflected in (22). At the temperatures and concentrations considered, we have $\sigma_{S3}\ll\sigma_{SL}$ in view of the "extra" factor $(\rho/\Delta\rho)^2\sim10^2$.

Note also there is also one more mechanism, not accounted for in the theoretical parts of Refs. 9 and 10, for heat transfer to the impurity subsystem. This mechanism is connected with Čerenkov emission of second-sound quanta by the thermal crystallization oscillations of the interface and by the Rayleigh waves, and is similar to the contribution, considered by Khalatnikov,[22] of the Rayleigh waves to the thermal resistance on an He II–metal interface. In the temperature region considered, the thermal crystallization waves with $\omega\sim T/\hbar\sim10^{10}$ s^{-1} are estimated to have an acoustic spectrum ($\omega\sim v_Mk$). The contribution of this mechanism to σ_{S3} therefore has the same temperature and concentration dependences as the contribution from the bulk phonons of the solid, viz., $\sigma^M_{S3}\propto c_2T^{7/2}(\rho/\Delta\rho)^2$. The exact expression for this contribution to σ_{S3} for $v_M\gg u_{II}$ is

$$\sigma_{S3}{}^M=\frac{\pi^3}{4}\left(\frac{\rho}{\Delta\rho}\right)^2\left(\frac{3}{5}m^*T\right)^{1/2}c_2\left(\frac{T}{\hbar v_M}\right)^3,$$

which is also of the same order as the results of Refs. 9 and 10. We note in conclusion that the impurity part of the interface thermal conductivity also receives a contribution from Čerenkov emission of Rayleigh waves. This contribution is $\sigma^R_{S3}\propto c_2T^{7/2}(\rho/\Delta\rho)^2$ just as for crysallization waves, and has the same order of magnitude.

We are grateful to A. F. Andreev for support and help. We are indebted to Yu. D. Anufriev, E. P. Bashkin, and A. É. Meĭerovich for helpful discussions and advice, and also to K. N. Zinov'eva, A. V. Markelov, and A. P. Sebedash for their interest in the work.

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Translated by J. G. Adashko