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Fluctuation subharmonic and multiharmonic phonon transmission and Kapitza conductance between crystals with very different vibrational spectra

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The existence of low-frequency, almost dispersionless intrinsic resonant vibrational modes in a transition layer at clean interface between two media (a rigid and a soft crystal) with very different elastic properties is predicted. It is shown that nonlinear interaction of bulk acoustic phonons with fluctuation resonant vibrations of a two-dimensional transition layer leads to subharmonic transmission across the interface of acoustic phonons incident from a rigid crystal with frequencies above the highest frequency in a soft crystal, and to the second- (and multiple-) harmonic transmission of acoustic phonons incident from a soft crystal. It is shown that contrary to subharmonic and multiharmonic generation of coherent acoustic waves, the coefficients of fluctuation subharmonic and multiharmonic phonon transmission do not depend on the amplitude of incident acoustic wave and are determined by temperature-dependent mean-square amplitudes of relative interface fluctuation displacements. It is emphasized that fluctuation subharmonic phonon transmission is a nonthreshold dynamical phenomenon which is significant for its contribution to Kapitza thermal boundary conductance across the interface. In both cases of fluctuation subharmonic and multiharmonic phonon transmission, two-dimensional resonant interface transition layer leads to the considerable enhancement of the coupling between phonons in the media with very different elastic properties. It is shown that inelastic interface dynamical phenomena can substantially contribute at elevated temperatures to the Kapitza conductance between solids with very different vibrational spectra (such as diamond and soft crystal) and to the thermal conductance across a helium-solid interface also.

It is well known from the linear wave theory that the transmission of the bulk acoustic waves across a sharp interface of two materials is strongly suppressed in the case of large differences in their elastic properties (e.g., density and sound velocity). If applied to thermal phonons, this phenomenon leads to Kapitza thermal boundary resistance at the interface between liquid helium (^{3}He and ^{4}He) and a wide variety of solids, and at the interface between two different solids also.[1–3] However, in the case of a helium-solid interface the acoustic-mismatch phonon theory of the Kapitza resistance[1] appears to be adequate only for very low temperatures (usually below 0.1 K). Experimental values of the Kapitza conductance at elevated temperatures are much larger (by a factor of 10 or 100) than estimated by this approach and no satisfactory explanation of this long-standing disagreement has been given.[2,3] The solidification of helium also does not substantially change the transmission of the phonons across a helium-solid interface,[3] and Kapitza thermal boundary conductance to solid helium,[4] as well as to solid hydrogen and deuterium,[5] is anomalously large. In a recent paper[6] measurements of Kapitza thermal boundary conductance between diamond and several metals were reported. Such interfaces represent the interfaces between solids with widely differing elastic properties because diamond has the highest sound velocity and Debye temperature of any material. In these experiments it was revealed that at the interface between diamond and such soft metals with very low Debye temperatures as Au and Pb, the Kapitza conductances measured at room temperatures are as much as 100 times larger than expected from the conventional acoustic-mismatch phonon theory. From these observations it follows that there is some additional way, intrinsic to diamond, in which energy is transferred across the interface. It implies, in particular, that phonons with frequencies above the highest frequency in soft metal must substantially contribute to the energy transfer.[6]

In a large number of works on the problem of Kapitza thermal boundary conductance (see Refs. 2,3 and references therein), the importance was revealed of the interface transition layer which gives rise to a better matching between phonons in two media. A two-dimensional transition layer can cause, in particular, an anomalous resonant absorption of incident bulk acoustic waves[7] and a total resonant transmission of phonons across the interface between two crystals with very different elastic properties[8] (see also Refs. 9–11). In the present paper, on the ground of rather general considerations it is shown that intrinsic resonant modes with frequencies within the vibrational continuum of a soft crystal can exist in the transition layer at the clean interface between two solids (a rigid and a soft crystal) with very different elastic properties. The origin of these modes lies in a structure of several atomic layers of a soft crystal adjacent to a rigid crystal. The strengths of the interlayer bonds, holding a soft crystal to a rigid one, are in the general case larger than the bulk interlayer bonds in a soft crystal.[12–16] Therefore the first (two or three) atomic layers of a soft crystal, adjacent to a rigid one, are strongly compressed with respect to the bulk layers of a soft crystal and the corresponding interlayer force constants in the layers are larger than the bulk ones in a soft crystal. (In the case of a helium-solid interface it leads to the solidification of the first two or three layers of helium, adsorbed on a solid, even in the case when the system is not under ambient pressure; see, e.g., Ref. 16.) It means that the first several atomic layers of a soft crystal form one "heavy transition layer" which is characterized by the existence of intrinsic resonant modes with relatively low frequencies. These resonant modes are the lowest transversely quantized standing-wave vibrational modes confined in the layer (see, e.g., Ref. 7). Their frequencies are determined by the strengths of the bonds with a rigid crystal of the first atomic layer of a soft crystal, adjacent to a rigid one, and by total thickness of the transition layer (which consists of several atomic layers of a soft crystal). Low-frequency surface resonant modes can exist at the interface between helium (liquid or solid) and an ordinary solid, and also at the interface between diamond and a soft crystal (such as Au or Pb). Low-damping almost dispersionless surface excitations were found by inelastic neutron scattering in the layers of ^{4}He adsorbed on different solid substrates.[16] The origin of these surface excitations may be the first two or three solidified helium layers, adjacent to the solid, formed due to the substrate potential.[16] A "breakdown" of the acoustic phonon reflectivity at a helium-solid interface was observed roughly at the same frequencies.[17] A strong phonon conversion due to only three atomic layers of helium at the interface with a solid was also observed experimentally,[18] which testifies that inelastic interface processes play a significant role in the phenomenon of anomalous Kapitza thermal boundary conductance.

In the present paper it is shown that the nonlinear interaction of bulk acoustic phonons with *fluctuation* resonant vibrations of a two-dimensional transition layer leads to several inelastic interface dynamical phenomena: (1) to subharmonic transmission across the interface of acoustic phonons incident from a rigid crystal with frequencies *above* the highest frequency in a soft crystal and (2) to second- (and multiple-) harmonic transmission across the interface of acoustic phonons incident from a soft crystal due to the interface generation of acoustic vibrations in a rigid crystal with frequencies *multiple* to resonant frequencies. It is shown that contrary to subharmonic and multiharmonic generation of coherent acoustic waves, the coefficients of fluctuation subharmonic and multiharmonic phonon transmission do not depend on the amplitude of the incident acoustic wave and are determined by temperature-dependent mean-square amplitudes of relative interface fluctuation displacements. It is emphasized that fluctuation subharmonic phonon transmission is a *nonthreshold* dynamical phenomenon which is significant for its contribution to Kapitza thermal boundary conductance across the interface. In both cases of fluctuation subharmonic and multiharmonic phonon transmission, a two-dimensional resonant interface transition layer leads to considerable enhancement of the coupling between phonons in the media with very different elastic properties. A substantial contribution at elevated (room) temperatures of inelastic interface phenomena to the Kapitza conductance between very different solids (such as diamond and soft crystal like Pb or Au) is estimated. The origin of the described inelastic interface dynamical processes is different from previously considered nonresonant high-frequency "bulk" three-phonon processes near the interface between ordinary and quantum crystals.[19] The proposed nonlinear dynamical model does not imply a significant electronic contribution to anomalous Kapitza conductance across the interface[20] since such a contribution was not confirmed experimentally.[6] A brief description of the nonlinear interaction of bulk acoustic waves with resonant vibrations of a two-dimensional transition layer and its contribution to Kapitza thermal boundary conductance was done in Ref. 21.

For a consistent macroscopic description of the low-frequency dynamical properties of a two-dimensional resonant layer it is necessary to account for the discontinuity (on the interatomic scale) of the elastic displacements at the interface between two bonded crystals (see, e.g., Refs. 22 and 23). A similar approach has been already used for the macroscopic description of the long-wavelength dynamics of an adsorbed monolayer weakly bonded with a crystal surface.[24,25] When the strengths of the interlayer bonds in the transition layer are intermediate between the strengths of the bulk bonds in the adjacent crystals, the transition layer is strongly bonded to a soft crystal but weakly bonded to a rigid one. Therefore a center-of-mass displacement $\vec{u}^s$ of the transition layer (placed at the plane $z=0$) coincides with the edge displacement $\vec{u}_2(z=0)\equiv\vec{u}_2(0)$ of a soft crystal (subscript 2) [i.e., $\vec{u}^s=\vec{u}_2(0)$], but does not coincide with the edge displacement $\vec{u}_1(0)$ of a rigid crystal (subscript 1). If we consider the (001) interface between two (cubic) crystals, the surface part of the Lagrangian function of the macroscopic elastic motion will have the following two-dimensional density $\mathcal{L}_s$:

$$\begin{aligned}\mathcal{L}_s &= \tfrac{1}{2}\sum_{i=1}^{3}\rho_s(\dot{u}_i^s)^2-\mathcal{U},\\ \mathcal{U} &= d_z\Delta_z+\tfrac{1}{2}A_{xx}(\Delta_x^2+\Delta_y^2)+\tfrac{1}{2}A_{zz}\Delta_z^2+B_{zxx}\Delta_z(\Delta_x^2+\Delta_y^2)+\tfrac{1}{3}B_{zzz}\Delta_z^3\\ &\quad+\tfrac{1}{4}C_{xxxx}(\Delta_x^4+\Delta_y^4)+\tfrac{1}{2}C_{xxyy}\Delta_x^2\Delta_y^2+\tfrac{1}{2}C_{xxzz}\Delta_z^2(\Delta_x^2+\Delta_y^2)+\tfrac{1}{4}C_{zzzz}\Delta_z^4. \end{aligned}\tag{1}$$

Here ρ_s is the surface mass per unit area of the interface, $\Delta_i\equiv u_i^s-u_{i,1}(0)=u_{i,2}(0)-u_{i,1}(0)$ is a pseudovector describing the relative interface displacements (surface displacement discontinuity),[22] A_{ik} and C_{iklm} are the tensors of the harmonic and quartic anharmonic force constants, B_{ikl} is a pseudotensor of the cubic anharmonic force constants describing the *interlayer* interaction between the transition layer and a rigid crystal, and d_i is a macroscopic pseudovector parameter describing the equilibrium surface (interface) relaxation Δ_{0i} in a soft crystal 2; see Eq. (5) below. [The latter describes the change of the equilibrium interlayer spacing, with respect to the bulk one in a soft crystal, between the adjacent atomic planes of soft and rigid crystals. Surface relaxation occurs both at the free surfaces of crystals (see, e.g., Ref. 26) and at interfaces between different crystals (see, e.g., Ref. 27).] In Eq. (1) it is taken into account that the d_z, $B_{zxx} = B_{zyy}$, and B_{zzz} components of the odd-order pseudotensors d_i and B_{ikl} are in the general case *nonzero* at the considered two-dimensional interface layer due to the locally broken inverse symmetry along the normal to the interface even in the case when both adjacent crystals are centrosymmetric ones. (The OZ axis is directed along the normal to the interface from crystal 1 to crystal 2.) In Eq. (1) we neglect the lateral (intralayer) interface interaction (which is described by the interface elastic moduli and residual interface stress; see, e.g., Refs. 22,23) and the interaction between the interlayer and intralayer deformations, as not essential for the considered dynamical phenomena.

Introducing the atomic mass m_2 in a soft crystal and the nearest-neighbor harmonic and anharmonic interatomic force constants in the transition layer, we can estimate the macroscopic parameters of a two-dimensional transition layer given by Eq. (1):

$$\rho_s \sim N m_2 a^{-2}, \quad A \sim K^s a^{-2}, \quad B \sim L^s a^{-2}, \quad C \sim M^s a^2. \tag{2}$$

Here N is the number of soft-crystal atomic layers in a "heavy transition layer" (we assume that $N \simeq$ 2–3), K^s, L^s, and M^s are the nearest-neighbor harmonic and (cubic and quartic) anharmonic force constants between the atoms of the transition layer and a rigid crystal, and a is the equilibrium interatomic spacing in the transition layer. In the following we assume that, as was discussed above, the harmonic force constant K^s in the transition layer is intermediate between the force constants K_1 and K_2 in the adjacent crystals and that the elastic impedance of a rigid crystal considerably exceeds the impedance of a soft one: $K_1 \gg K^s > K_2$, $m_1 K_1 \gg m_2 K_2$.

In order to take into account the intrinsic dissipation in the two-dimensional interface layer, the surface dissipative function with density $\mathcal{R}_s$ per unit area should be introduced in addition to the surface Lagrangian function (cf. Ref. 7):

$$\mathcal{R}_s = \tfrac{1}{2}\Gamma_{xx}[(\dot{\Delta}_x)^2 + (\dot{\Delta}_y)^2] + \tfrac{1}{2}\Gamma_{zz}(\dot{\Delta}_z)^2. \tag{3}$$

The boundary conditions for bulk elastic stresses $\sigma_{ik}(0)$ and displacements $u_i(0)$ at the interface plane $z = 0$, which describe the macroscopic dynamics of a two-dimensional transition layer, can be obtained from the joint variation of the surface Lagrangian and dissipative functions and bulk elastic energy with respect to the *independent* variables $\vec{u}_1(0)$ and $\vec{u}_2(0) = \vec{u}^s$:

$$\sigma_{zi,1}(0) = -\frac{\delta \mathcal{L}_s}{\delta u_{i,1}} - \frac{\delta \mathcal{R}_s}{\delta \dot{u}_{i,1}}, \quad \sigma_{zi,2}(0) = \frac{\delta \mathcal{L}_s}{\delta u_{i,2}} + \frac{\delta \mathcal{R}_s}{\delta \dot{u}_{i,2}}, \quad \frac{\delta \mathcal{L}_s}{\delta u_i^s} + \frac{\delta \mathcal{R}_s}{\delta \dot{u}_i^s} = 0. \tag{4}$$

From Eqs. (1) and (4) and the requirements $\sigma_{zi,1}(0) = 0$, $\sigma_{zi,2}(0) = 0$ we obtain the equation for the uniform (along the interface) *equilibrium* static surface relaxation Δ_{0z} in a soft crystal 2:

$$C_{zzzz}\Delta_{0z}^3 + B_{zzz}\Delta_{0z}^2 + A_{zz}\Delta_{0z} = -d_z, \quad \Delta_{0x} = \Delta_{0y} = 0. \tag{5}$$

Since the surface relaxation Δ_{0z} is assumed to be less than the equilibrium interatomic spacing ($\Delta_{0z} \ll a$), the parameter d_z should be relatively small and the surface relaxation is described by the smallest root of Eq. (5): $\Delta_{0z} \approx -d_z/A_{zz}$.

Equations (4) with the use of Eqs. (1), (3), and (5) can be reduced to the following boundary conditions at $z = 0$ for the *dynamical* elastic fields u_i and $\Delta_i' = \Delta_i - \Delta_{0i}$ (near the equilibrium positions $\vec{u} = 0$ in the bulk of the adjacent crystals and $\Delta_{0x} = \Delta_{0y} = 0$ and finite Δ_{0z} at the interface plane $z = 0$):

$$\sigma_{zi,2} = \sigma_{zi,1} + \rho_s \ddot{u}_i^s, \tag{6}$$

$$\sigma_{zx,1} = [A_{xx} + 2B_{zxx}(\Delta_{0z} + \Delta_z') + C_{xxzz}(\Delta_{0z} + \Delta_z')^2]\Delta_x' + C_{xxxx}\Delta_x'^3 + C_{xxyy}\Delta_x'\Delta_y'^2 + \Gamma_{xx}\dot{\Delta}_x, \tag{7}$$

$$\sigma_{zy,1} = [A_{xx} + 2B_{zxx}(\Delta_{0z} + \Delta_z') + C_{xxzz}(\Delta_{0z} + \Delta_z')^2]\Delta_y' + C_{xxxx}\Delta_y'^3 + C_{xxyy}\Delta_y'\Delta_x'^2 + \Gamma_{xx}\dot{\Delta}_y, \tag{8}$$

$$\begin{aligned}\sigma_{zz,1} = {} & [A_{zz} + 2B_{zzz}\Delta_{0z} + 3C_{zzzz}\Delta_{0z}^2]\Delta_z' + [B_{zzz} + 3C_{zzzz}\Delta_{0z}]\Delta_z'^2 + C_{zzzz}\Delta_z'^3 \\ & + [B_{zxx} + C_{xxzz}(\Delta_{0z} + \Delta_z')](\Delta_x'^2 + \Delta_y'^2) + \Gamma_{zz}\dot{\Delta}_z.\end{aligned} \tag{9}$$

These boundary conditions consistently describe the low-frequency dynamics of the transition layer and account both for the interface discontinuity of elastic stresses [due to surface mass ρ_s; see Eqs. (6)] and for the interface discontinuity of elastic displacements $\Delta_i \equiv u_{i,2}(0) - u_{i,1}(0)$, $u_i^s = u_{i,2}(0)$ [due to the weak interlayer interaction at the interface; see Eqs. (7)–(9)]. More general boundary conditions can also be obtained within the framework of the macroscopic approach when the elastic displacement $\vec{u}^s$ of the transition layer coincides neither with $\vec{u}_1(0)$ nor with $\vec{u}_2(0)$ and two independent relative interface displacements $\Delta_i^{(1)} \equiv u_i^s - u_{i,1}(0)$ and $\Delta_i^{(2)} \equiv u_{i,2}(0) - u_i^s$ should be introduced (the corresponding linear boundary conditions are described in Ref. 23). Due to the interface anharmonic force constants B_{ikl} and C_{iklm}, the surface relaxation Δ_{0z} [Eq. (5)] accomplishes a *static renormalization* of the parameters A_{xx}, A_{zz}, B_{zxx}, and B_{zzz} in the interface equations of motion (7)–(9). Such a renormalization can be essential in the general case (and can result in the so-called "supermodulus effect" in some bimetal superlattices; see, e.g., Ref. 27). Therefore in the following we introduce renormalized parameters, namely,

$$\begin{aligned} & A_{xx}^* = A_{yy}^* = A_{xx} + 2B_{zxx}\Delta_{0z} + C_{xxzz}\Delta_{0z}^2, \quad A_{zz}^* = A_{zz} + 2B_{zzz}\Delta_{0z} + 3C_{zzzz}\Delta_{0z}, \\ & B_{zxx}^* = B_{zxx} + C_{xxzz}\Delta_{oz}, \quad B_{zzz}^* = B_{zzz} + 3C_{zzzz}\Delta_{0z}. \end{aligned} \tag{10}$$

With the use of these parameters, the boundary conditions (7)–(9) can be written in a more concise form:

$$\sigma_{zi,1} = A^*_{ik}\Delta'_k + B^*_{ikl}\Delta'_k\Delta'_l + C_{iklm}\Delta'_k\Delta'_l\Delta'_m + \Gamma_{ik}\dot{\Delta}_k. \tag{11}$$

To describe the low-frequency interface resonant mode polarized in the boundary plane, we look for the solution of bulk and interface equations of motion in the following form:

$$u_{x1} = u_{0x1}\exp\left(-i\frac{\omega}{c_{t1}}z - i\omega t\right),$$
$$u_{x2} = u_{0x2}\exp\left(i\frac{\omega}{c_{t2}}z - i\omega t\right),$$
$$u_{xs} = u_{0x2}\exp(-i\omega t), \tag{12}$$

where ρ and $c_{l,t}$ are the bulk densities and (longitudinal and transverse) sound velocities in the adjacent crystals. The above mode has a zero parallel to the interface component of the wave vector $\vec{k}$: $k_{\parallel} = 0$. Using the boundary conditions (6), (10), and (11) (in the linear case $A^* \gg B^*\Delta'$, $A^* \gg C^*\Delta'^2$), we ascertain that the low-frequency resonant mode with the following dispersion relation and the form of the displacement distribution exists at the interface between crystals with very different elastic properties:

$$\rho_s\omega^2 \simeq A^*_{xx} - i\omega\left(Z_{2t} + \frac{Z_{0t}^2}{Z_{1t}} + \Gamma_{xx}\right), \tag{13}$$

$$\frac{u_{0x1}}{u_{0x2}} \simeq i\frac{Z_{0t}}{Z_{1t}} \ll i, \tag{14}$$

where $Z_{0l,t} = \sqrt{A^*_{zz,xx}\rho_s}$ and $Z_{l,t} = \rho c_{l,t}$ are the longitudinal and transverse elastic impedances of the interface layer and the adjacent crystals. From Eq. (13) it follows that in the considered case when

$$Z_2 \ll Z_0 \ll Z_1, \quad Z_0^2 \ll Z_1 Z_2, \tag{15}$$

and $\rho_s > \rho_2 a$, $A^*_{xx}/\rho_s < (\pi^2\mu_2)/(\rho_2 a^2)$, the resonant frequency $\omega_{0\parallel} = \sqrt{A^*_{xx}/\rho_s} \approx \sqrt{K^s/Nm_2}$ is lower than the upper cutoff (Debye) frequency of a soft crystal, $\omega_{2\max} \approx \sqrt{K_2/m_2}$, and the damping of the interface resonant (pseudosurface) mode is caused mainly by the emission of bulk transverse elastic waves in a soft crystal 2 and by the intrinsic interface dissipation (which also finally results in the emission of noncoherent thermal phonons in a soft crystal to which the transition layer is strongly bonded). Equation (14) shows that the resonant mode is indeed accompanied by large *dynamical* relative interface displacements $\vec{\Delta}'_0 \equiv \vec{u}_{02} - \vec{u}_{01} \approx \vec{u}_{02}$. A similar conclusion follows also from the consideration of the discrete-lattice equations of motion in a simple one-dimensional model of the interface when the nearest-neighbor harmonic force constants $K^s_{n,n+1}$ (between the adjacent n and $n+1$ atomic layers in the transition layer) are different from the bulk force constants in a soft crystal at least in the first two near-surface layers, namely, $K_1 \gg K^s_{0,1} > K^s_{1,2} > K_2$ and $N = 2$ in relations (2).

With the use of boundary conditions (6), (10), and (11), we can show that the low-frequency *stretch* interface resonant mode polarized along the normal to the boundary also exists in the system at frequency $\omega_{0\perp} = \sqrt{A^*_{zz}/\rho_s}$. In the case of $A^*_{xx} = A^*_{yy}$ and uniform interface vibrations (with $k_{\parallel} = 0$), the two modes polarized in the interface plane have the same resonant frequency $\omega_{0\parallel} = \sqrt{A^*_{xx}/\rho_s}$. For surface modes with nonzero $k_{\parallel}$, the lateral interface elastic moduli delete the degeneracy between the frequencies of longitudinal and shear surface modes and cause a *small dispersion* of the modes (see, e.g., Ref. 8).

To consider a subharmonic acoustic phonon transmission across the interface, we assume that the double-resonant frequencies $2\omega_{0\parallel}$ and $2\omega_{0\perp}$ are higher than the upper cutoff frequency $\omega_{2\max}$ of a soft crystal 2. (Otherwise we can consider a subharmonic transmission of acoustic phonons with a near-triple-resonant frequency $\omega \simeq 3\omega_0$; see below.) In the linear system the incident from a rigid crystal acoustic phonon with near-double-resonant frequency $\omega \simeq 2\omega_0$ will be totally reflected from the interface. But at the interface *fluctuation* vibrations, either zero temperature or thermally excited, of the transition layer with near-resonant frequencies always have nonzero temperature-dependent amplitudes of the relative interface displacements $\Delta^f_{0i} = \Delta^f_{0i}(T)$. Since we assume that low-frequency interface resonant modes are almost dispersionless and, according to Eq. (14), one has $\vec{\Delta}'_0 \approx \vec{u}_{02}$ in these modes, the temperature dependences of the mean-square amplitudes $\langle\Delta^f_{0i}(T)^2\rangle$ of relative interface fluctuation displacements can be described [in the small-amplitude limit $\langle\Delta^f_{0i}(T)^2\rangle \ll a^2$] in a model of a two-dimensional array of (anisotropic) Einstein oscillators:

$$\langle\Delta^f_{0x}(T)^2\rangle = \langle\Delta^f_{0y}(T)^2\rangle = \frac{\hbar}{2Nm_2\omega_{0\parallel}}\coth\left(\frac{\hbar\omega_{0\parallel}}{2k_BT}\right), \quad \langle\Delta^f_{0z}(T)^2\rangle = \frac{\hbar}{2Nm_2\omega_{0\perp}}\coth\left(\frac{\hbar\omega_{0\perp}}{2k_BT}\right), \tag{16}$$

where N is the number of soft-crystal atomic layers in a "heavy transition layer" [see Eq. (2)].

The anharmonic interaction of incident acoustic waves (with near-double-resonant frequency ω) with fluctuation resonant interface vibrations is described by the term $B^*_{ikl}\Delta'_k(\omega)\Delta^f_l(\omega_0)$ in right-hand side (rhs) of Eqs. (11) and results in the interface stress $\sigma_{zi,1}$ with near-resonant frequency. Therefore the incident from a rigid crystal high-frequency phonons excite the interface oscillations with near-resonant frequencies which in turn emit phonons in a soft crystal. In the inverse inelastic process of second- (or multiple-) harmonic generation, the incident soft-crystal phonons with near-resonant frequencies excite the interface oscillations with near-double- (and near-multiple-) resonant frequencies which emit high-frequency phonons in a rigid crystal.

To describe in the main approximation the subharmonic acoustic phonon transmission, we have to solve the linear boundary problem and find the relative interface displacements $\Delta'_k(\omega)$ induced by the incident wave. By solving Eqs. (6), (10), and (11) in the linear case in the assumption (15) for longitudinal rigid-crystal acoustic waves with amplitude u_1 normally incident at the interface, we obtain the amplitudes of reflected ru_1 and transmitted tu_1 waves as well as the amplitudes of the displacement of the transition layer $u^s \equiv tu_1$ and of relative interface displacement $\Delta_2 \equiv u^s - u_1(1+r)$:

$$r = \frac{Z_{1l}Z_{2l}\omega + i[Z_{1l}(A^{*\prime}_{zz} - \rho_s\omega^2) - Z_{2l}A^{*\prime}_{zz}]}{Z_{1l}Z_{2l}\omega + i[Z_{1l}(A^{*\prime}_{zz} - \rho_s\omega^2) + Z_{2l}A^{*\prime}_{zz}]}, \quad (17)$$

$$t \equiv \frac{u^s}{u_1} = \frac{2iZ_{1l}A^{*\prime}_{zz}}{Z_{1l}Z_{2l}\omega + i[Z_{1l}(A^{*\prime}_{zz} - \rho_s\omega^2) + Z_{2l}A^{*\prime}_{zz}]}, \quad (18)$$

$$\Delta_2 \equiv u^s - u_1(1+r) = u_1\frac{2Z_{1l}\omega(i\rho_s\omega - Z_{2l})}{Z_{1l}Z_{2l}\omega + i[Z_{1l}(A^{*\prime}_{zz} - \rho_s\omega^2) + Z_{2l}A^{*\prime}_{zz}]}, \quad (19)$$

where $A^{*\prime}_{ik} = A^*_{ik} - i\omega\Gamma_{ik}$.

From Eq. (18) we see that for *low-frequency* waves (with $\omega \ll \omega_{0\perp} < \omega_{2\max}$), the coefficient of acoustic power transmission across the interface is small, $T(\omega) = |t|^2 Z_{2l}/Z_{1l} \approx 4Z_{2l}/Z_{1l} \ll 1$, but we have a strong resonant enhancement of the coefficient for $\omega \approx \omega_{0\perp}$ (see also Ref. 8). For low-damping interface oscillations (when $A^{*\prime} \approx A^*$), the coefficient of *resonant harmonic* power transmission, $T^l_{\rm RH}(\omega)$, of longitudinal acoustic waves can be approximately written in the assumption (15) in the form of a δ function, which is convenient for the further calculation of phonon heat flux across the interface [see Eq. (24) below]:

$$T^l_{\rm RH}(\omega) = \frac{2\pi A^*_{zz}}{Z_{1l}}\delta(\omega - \omega_{0\perp}). \quad (20)$$

The coefficient $T^t_{\rm RH}(\omega)$ of resonant harmonic power transmission of transverse acoustic phonons is correspondingly equal to $[2\pi A^*_{xx}/Z_{1t}]\delta(\omega - \omega_{0\|})$. Using Eqs. (6), (10), and (11) in the linear case for the arbitrary ratio between Z_0^2 and Z_1Z_2 [instead of the assumption (15)], we can show that for given resonant frequencies the coefficients of resonant harmonic phonon transmission reach their maximal values of $T^{t,l}_{\rm RH}(\omega_{0\|,\perp}) = 1$ (when correspondingly $r = 0$) for $Z^2_{0t,l} = Z_{1t,l}Z_{2t,l}$. This resonant surface phenomenon is similar to the acoustic clearing of an interface between two media due to a macroscopic quarter-wavelength transition layer with acoustic impedance equal to a geometric mean of the impedances of the contacting media (see, e.g., Ref. 8).

In the *high-frequency* domain $\omega > \omega_{2\max}$, effective acoustic impedance of a soft crystal is a pure imaginary one $Z_{2l} = -iZ''_{2l}$ and Eq. (17) describes in this case (almost) total reflection of incident rigid-crystal acoustic waves, when $|r| \approx 1$ and in a soft crystal there are only evanescent high-frequency waves decaying into the bulk. To describe fluctuation subharmonic transmission of high-frequency phonons, we consider rigid-crystal longitudinal waves normally incident at the interface with amplitude u_1 and near-double-resonant frequency $\omega = 2\omega_{0\|} + \epsilon$ (ϵ is a small detuning). In the assumption $Z''_{2l} \ll Z_{1l}$ for the high-frequency dynamical impedance of a soft crystal, from Eq. (19) we find the relative interface displacements Δ_2 induced by the incident wave: $|\Delta_2/u_1| \approx 8/3$. With the same assumption we find that the interface stress with near-resonant frequency $\gamma = \omega_{0\|} + \epsilon$ is described by the term $\sigma_{zx,1} = B^*_{zxx}\Delta_2\Delta^f_{0x}\cos(\gamma t)$ in Eq. (7). This surface force is applied to the interface oscillator with the damping given by the imaginary part of Eq. (13). From the expression for the response of the oscillator to the driving force with near-resonant frequency,[28] we readily find the average absorption $I(\epsilon)$ (per unit time and unit area) of the acoustic energy by the interface oscillator:

$$I(\epsilon) \approx \frac{7B^{*2}_{zxx}u_1^2\langle\Delta^{f2}_{0x}\rangle}{4\rho_s}\frac{\lambda}{\epsilon^2 + \lambda^2}, \quad (21)$$

where $\lambda \approx (Z_2 + \Gamma_{xx})/(2\rho_s)$ is a parameter describing the damping of the oscillator [see Eq. (13)]. After the normalization of the interface absorption $I(\epsilon)$ by the flux of the energy $I_0 = u_1^2\omega^2 Z_1 \approx 4u_1^2\omega_0^2 Z_1$ in the incident rigid-crystal acoustic wave, we obtain the coefficient of the power transmission $T(\omega)$ which finally determines the heat flux due to the transmission of acoustic phonons from rigid crystal 1 to soft crystal 2. In the considered case of low-damping interface oscillations $\lambda \ll \omega_0$ (when $Z_2 + \Gamma_{xx} \ll Z_0$), from Eq. (21) it follows that the coefficient of fluctuation subharmonic transmission, $T^t_{\rm FSH}(\omega, T)$, of longitudinal rigid-crystal phonons into *transverse* waves in a soft crystal can be written in the form of a δ function:

$$T^t_{\rm FSH}(\omega, T) = \frac{7\pi B^{*2}_{zxx}\langle\Delta^f_{0x}(T)^2\rangle}{16A^*_{xx}Z_{1l}}\delta(\omega - 2\omega_{0\|}). \quad (22)$$

In the case of longitudinal acoustic phonons normally incident at the interface from a rigid crystal with frequency ω close to $2\omega_{0\perp}$, the coefficient of fluctuation subharmonic power transmission, $T^l_{\rm FSH}(\omega, T)$, into *longitudinal* waves in a soft crystal has a resonant form similar to Eq. (22):

$$T^l_{\rm FSH}(\omega, T) = \frac{7\pi B^{*2}_{zzz}\langle\Delta^f_{0z}(T)^2\rangle}{16A^*_{zz}Z_{1l}}\delta(\omega - 2\omega_{0\perp}). \quad (23)$$

Using Eqs. (6), (10), and (11) we can show that for *oblique* incidence at the interface of high-frequency rigid-crystal phonons, the corresponding coefficient of fluctuation subharmonic transmission also has a characteristic resonant form similar either to Eq. (22) or to Eq. (23).

In the case of rigid-crystal transverse (or longitudinal) phonons normally incident at the interface with near-triple-resonant frequency $\omega \simeq 3\omega_{0\|}$ (or with $\omega \simeq 3\omega_{0\perp}$), the coefficient of fluctuation subharmonic power transmission into transverse (or longitudinal) elastic waves in a soft crystal has a resonant form similar to Eqs. (22) and (23) and is proportional to $\{[C_{xxxx}\langle\Delta^f_{0x}(T)^2\rangle + (1/3)C_{xxyy}\langle\Delta^f_{0y}(T)^2\rangle]^2/(A^*_{xx}Z_{1t})\}\delta(\omega - 3\omega_{0\|})$ (or to $[C^{*2}_{zzzz}\langle\Delta^f_{0z}(T)^4\rangle/(A^*_{zz}Z_{1l})]\delta(\omega - 3\omega_{0\perp})$). Similar properties possess the coefficients of fluctuation subharmonic transmission of rigid-crystal phonons incident with higher-order near-multiple-resonant frequencies. It is important that, contrary to subharmonic generation of coherent acoustic waves, the coefficients of fluctuation subharmonic transmission do not depend on the amplitude of *incident* acoustic waves and fluctuation subharmonic phonon transmission is a *nonthreshold* dynamical phenomenon, which is significant for its contribution to Kapitza thermal boundary conductance across the interface.

The heat flux $q_1(T)$ (per unit area of the interface) due to the transmission of acoustic phonons from rigid crystal 1 to soft crystal 2 with the coefficient of power transmission $T_{\rm AP}(\omega, T)$ can be written as follows (see, e.g., Ref. 16):

$$q_1(T) = \frac{1}{2}\int_0^{\omega_{1\,\rm max}} n(\omega,T)D_1(\omega)\hbar\omega\langle v_z\rangle\langle T_{\rm AP}(\omega,T)\rangle d\omega. \tag{24}$$

Here $n(\omega, T)$ is the Bose-Einstein distribution function for phonons with frequency ω at temperature T, $D_1(\omega)$ is the density of phonon states in a rigid crystal 1, $\langle v_z\rangle$ and $\langle T_{\rm AP}(\omega,T)\rangle$ are the average (over the three acoustic phonon branches) values of the z components of the group velocities and coefficients of power transmission of the incident phonons. Kapitza thermal boundary conductance $\sigma_K(T)$ is proportional to the temperature derivative of the heat flux across the interface: $\sigma_K(T) \propto \partial q_1(T)/\partial T$.

In order to compare the contributions to the heat flux and, correspondingly, to the Kapitza conductance of fluctuation subharmonic and nonresonant harmonic phonon transmission, we have to take into account a steep dependence of the density of phonon states in a rigid crystal $[D_1(\omega) \propto \omega^2]$ in the low-frequency domain $\omega \ll \omega_{1\,\rm max}$ and that the coefficient $T_{\rm NRH}$ of nonresonant harmonic transmission of acoustic waves across a *sharp* interface between acoustically strongly mismatched crystals is of the order of $T_{\rm NRH} \simeq 4Z_1Z_2/(Z_1+Z_2)^2 \simeq 4Z_2/Z_1 \ll 1$ [see Eq. (18)]. For $T \geq 2\hbar\omega_0$ which (approximately) corresponds to room temperatures used in [6], from Eqs. (22)–(24) we can estimate the ratio between the heat flux $q_{1\rm SH}(T)$ due to fluctuation subharmonic (one-half-harmonic) transmission across the interface with a resonant transition layer of high-frequency rigid-crystal phonons (with $\omega \geq \omega_{2\,\rm max}$) and of the heat flux $q_{1\rm NRH}(T)$ due to nonresonant harmonic transmission across a sharp interface of low-frequency rigid-crystal phonons (with $\omega \leq \omega_{2\,\rm max} \ll \omega_{1\,\rm max}$):

$$\frac{q_{1\rm SH}(T)}{q_{1\rm NRH}(T)} \simeq \frac{B^{*2}\langle\Delta_0^f(T)^2\rangle}{AZ_2\omega_0}. \tag{25}$$

According to Eq. (16), this ratio increases with increasing temperature. For soft crystals with relatively low melting temperatures (such as crystals of Pb and Au) or quantum crystals of ^{3}He and ^{4}He, at elevated temperatures the mean-square amplitudes of relative interface fluctuation displacements can reach the large values of $\langle\Delta_0^f(T)^2\rangle \sim A^2/B^{*2}$ [when the harmonic-oscillators approximation (16) fails]. (Extremely large thermal vibrations of weakly bonded adsorbed atoms have been recently observed on crystal surfaces at elevated temperatures.[29]) In the case of large thermal vibrations of soft-crystal atoms in the interface layer, the ratio between fluctuation subharmonic and nonresonant harmonic heat fluxes (25) reaches the large value of $A/Z_2\omega_0 \simeq Z_0/Z_2 \gg 1$. At elevated temperatures the contribution to the heat flux $q_1(T)$ due to fluctuation subharmonic transmission of rigid-crystal phonons incident with near-triple-resonant frequencies $\omega \simeq 3\omega_0$ (and with higher-order near-multiple-resonant frequencies) is of the same order of magnitude as the contribution due to fluctuation one-half-harmonic phonon transmission. It means that the additional heat flux from a rigid to a soft crystal due to fluctuation subharmonic transmission of rigid-crystal phonons incident with frequencies *above* the highest frequency in a soft crystal increases with increasing ratio between Debye temperatures of rigid and soft crystals. (In the case of diamond and Pb or Au this ratio is approximately 25 or 14, respectively.[6]) At elevated temperatures the phonon heat flux $q_{1\rm RH}(T)$ due to *resonant* harmonic transmission [with the coefficient $T_{\rm RH}(\omega)$ given by Eq. (20)] of rigid-crystal phonons across the interface with the transition layer also has a relatively large value of $q_{1\rm RH}(T)/q_{1\rm NRH}(T) \simeq Z_0/Z_2 \gg 1$. Therefore due to fluctuation subharmonic and resonant harmonic phonon transmission, a two-dimensional interface transition layer can substantially contribute at elevated temperatures to the Kapitza conductance between crystals with very different vibrational spectra.

To describe the main features of inverse inelastic process of acoustic second-harmonic generation at a two-dimensional transition layer, we consider the normal incidence from a soft crystal of transverse acoustic waves with frequency $\omega < \omega_{2\,\rm max}$ and amplitude u_2. In this case we have reflected and transmitted transverse waves with frequency ω and transmitted longitudinal waves with double frequency 2ω and amplitude $u_{1,2}$. By solving in the assumption (15) the linear boundary problem for the waves with frequency ω, we find the amplitudes of reflected ru_2 and transmitted tu_2 waves as well as the amplitudes of the displacement of the transition layer $u^s \equiv (1+r)u_2$ and of the relative interface displacement $\Delta_1 \equiv u^s - u_2 t$ [cf. Eqs. (17)–(19)]:

$$r = \frac{Z_{1t}Z_{2t}\omega + i[Z_{2t}A^{*\prime}_{xx} - Z_{1t}(A^{*\prime}_{xx} - \rho_s\omega^2)]}{Z_{1t}Z_{2t}\omega + i[Z_{2t}A^{*\prime}_{xx} + Z_{1t}(A^{*\prime}_{xx} - \rho_s\omega^2)]}, \tag{26}$$

$$t = \frac{2iZ_{2t}A^{*\prime}_{xx}}{Z_{1t}Z_{2t}\omega + i[Z_{2t}A^{*\prime}_{xx} + Z_{1t}(A^{*\prime}_{xx} - \rho_s\omega^2)]}, \tag{27}$$

$$u^s = u_2\frac{2Z_{2t}(Z_{1t}\omega + iA^{*}_{xx})}{Z_{1t}Z_{2t}\omega + i[Z_{2t}A^{*\prime}_{xx} + Z_{1t}(A^{*\prime}_{xx} - \rho_s\omega^2)]}, \tag{28}$$

$$\Delta_1 \equiv u^s - u_1 t = u_2\frac{2Z_{11}Z_{2t}\omega}{Z_{1t}Z_{2t}\omega + i[Z_{2t}A^{*\prime}_{xx} + Z_{1t}(A^{*\prime}_{xx} - \rho_s\omega^2)]}. \tag{29}$$

From Eq. (27) we see that for *low-frequency* waves (with $\omega \ll \omega_{0\|} < \omega_{2\,\rm max}$), the coefficient of harmonic acoustic power transmission across the interface is small, $T(\omega) = |t|^2 Z_{1t}/Z_{2t} \approx 4Z_{2t}/Z_{1t} \ll 1$, but we have a strong resonant enhancement of the coefficient for $\omega \approx \omega_{0\|}$. For low-damping interface oscillations, the coefficient $T^t_{\rm RH}(\omega)$ of *resonant harmonic* power transmission of transverse waves can be approximately written in the assumption (15) in the form of a δ function, similar to Eq. (20):

$$T^t_{\rm RH}(\omega) = \frac{2\pi A^*_{xx}}{Z_{1t}}\delta(\omega - \omega_{0\parallel}). \tag{30}$$

The coefficient $T^l_{\rm RH}(\omega)$ of resonant harmonic power transmission of longitudinal phonons is correspondingly equal to $[2\pi A^*_{zz}/Z_{1l}]\delta(\omega - \omega_{0\perp})$.

From Eqs. (9) and (29) in the assumption (15) we find, for near-resonant frequencies $\omega = \omega_{0\parallel} + \epsilon$ the amplitude of relative interface displacements Δ_1 and generating interface stress $\sigma_{zz,1}$ for second-harmonic longitudinal waves in crystal 1,

$$\Delta_1^2 = \frac{4u_2^2}{1 + (2\rho_s\epsilon/Z_2)^2}, \tag{31}$$

$$\sigma_{zz,1} = \tfrac{1}{2}B^*_{zxx}\Delta_1^2\cos(2\omega t). \tag{32}$$

Since $u_{1,2} \approx |\sigma_{zz,1}|/(2\omega Z_{1l})$, from Eqs. (31) and (32) we find the form of the coefficient $T^t_{\rm RDH}(\omega, T)$ of *resonant* interface power transmission of transverse soft-crystal phonons into longitudinal second-harmonic acoustic waves in a rigid crystal:

$$T^t_{\rm RDH}(\omega) = \frac{4Z_{1l}u_{1,2}^2}{Z_{2t}u_2^2} \approx \frac{\pi B^{*2}_{zxx}u_2^2}{A_{xx}Z_{1l}}\delta(\omega - \omega_{0\parallel}). \tag{33}$$

In the case of longitudinal acoustic phonons normally incident from a soft crystal with amplitude u_2 and frequency ω close to $\omega_{0\perp}$, the coefficient $T^l_{\rm RDH}(\omega)$ of resonant power transmission into longitudinal second-harmonic acoustic waves in a rigid crystal has a resonant form similar to Eq. (33) and is (approximately) equal to $[\pi B^{*2}_{zzz}u_2^2/(A_{zz}Z_{1l})]\delta(\omega - \omega_{0\perp})$. In both cases the coefficients of resonant second-harmonic transmission are proportional to the square of the amplitude of incident soft-crystal acoustic waves.

Another channel of second-harmonic phonon transmission is determined, as in the case of subharmonic transmission, by the interaction of incident soft-crystal acoustic waves with *fluctuation* resonant vibrations of the transition layer. Fluctuation generating interface stress for second-harmonic elastic waves in crystal 1 is determined by Eq. (11), instead of Eq. (31), as follows:

$$\sigma_{zz,1} = B^*_{zxx}\Delta_1\Delta^f_{0x}\cos(2\omega t). \tag{34}$$

From Eqs. (31), (33), and (34) in the assumption (15) we find the coefficient $T^t_{\rm FDH}(\omega, T)$ of *fluctuation* second-harmonic transmission of *transverse* soft-crystal phonons into longitudinal waves in a rigid crystal:

$$T^t_{\rm FDH}(\omega, T) = \frac{2\pi B^{*2}_{zxx}\langle\Delta^f_{0x}(T)^2\rangle}{A^*_{xx}Z_{1l}}\delta(\omega - \omega_{0\parallel}). \tag{35}$$

The coefficient $T^l_{\rm FDH}(\omega, T)$ of fluctuation second-harmonic transmission of *longitudinal* soft-crystal phonons into longitudinal waves in a rigid crystal has a resonant form similar to Eq. (35):

$$T^l_{\rm FDH}(\omega, T) = \frac{2\pi B^{*2}_{zzz}\langle\Delta^f_{0z}(T)^2\rangle}{A^*_{zz}Z_{1l}}\delta(\omega - \omega_{0\perp}). \tag{36}$$

The coefficient of fluctuation *third-harmonic* power transmission of near-resonant transverse (or longitudinal) soft-crystal phonons into elastic waves in a rigid crystal also has a resonant form and is proportional to $\{[C_{xxxx}\langle\Delta^f_{0x}(T)^2\rangle + (1/3)C_{xxyy}\langle\Delta^f_{0y}(T)^2\rangle]^2/(A^*_{xx}Z_{1t})\}\delta(\omega - \omega_{0\parallel})$ (or to $[C^{*2}_{zzzz}\langle\Delta^f_{0z}(T)^4\rangle/(A^*_{zz}Z_{1l})]\delta(\omega - \omega_{0\perp})$). properties possess the coefficients of fluctuation *multiharmonic* transmission of soft-crystal phonons across the interface. It is important that in all cases the coefficients of fluctuation multiharmonic transmission do not depend on the amplitude of incident soft-crystal acoustic waves.

From the comparison of Eqs. (35),(36) and (22),(23) we see that the coefficients $T^{l,t}_{\rm FDH}(\omega, T)$ of fluctuation second-harmonic phonon transmission are similar to the coefficients $T^{l,t}_{\rm FSH}(\omega, T)$ of fluctuation subharmonic (one-half-harmonic) phonon transmission which reflects the *reciprocity* of these inelastic interface dynamical processes and their contribution to the phonon heat flux across the interface.

In conclusion, the existence of low-frequency almost dispersionless intrinsic resonant vibrational modes in the transition layer at the clean interface between two media (rigid and soft crystals) with very different elastic properties is predicted. It is shown that nonlinear interaction of bulk acoustic phonons with *fluctuation* resonant vibrations of a two-dimensional transition layer leads to subharmonic transmission across the interface of acoustic phonons incident from a rigid crystal with frequencies *above* the highest frequency in a soft crystal, and to second- (and multiple-) harmonic transmission of acoustic phonons incident from a soft crystal. It is shown that, contrary to subharmonic and multiharmonic generation of coherent acoustic waves, the coefficients of fluctuation subharmonic and multiharmonic phonon transmission do not depend on the amplitude of incident acoustic waves and are determined by temperature-dependent mean-square amplitudes of relative interface fluctuation displacements. It is emphasized that fluctuation subharmonic phonon transmission is a *nonthreshold* dynamical phenomenon which is significant for its contribution to Kapitza thermal boundary conductance across the interface. In both cases of fluctuation subharmonic and multiharmonic phonon transmission, a two-dimensional resonant interface transition layer leads to considerable enhancement of the coupling between phonons in the media with very different elastic properties. A substantial contribution at elevated (room) temperatures of inelastic interface phenomena to the Kapitza conductance between very different solids (such as diamond and soft crystals like Pb or Au) is considered. The existence of low-frequency resonant modes at the clean interface between crystals with very different vibrational spectra can be verified by infrared-absorption[30] or Raman-scattering[31] spectroscopies which have already been used for the investigation of two-dimensionally localized and planar vibrational modes at buried monatomic layers and interfaces.

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