Effects of isotropic overlayer on the focusing of surface phonons

T. Aono and S. Tamura
Department of Applied Physics, Hokkaido University, Sapporo 060, Japan
(Received 12 August 1996)

Based on the geometrical acoustic approximation, we study theoretically the phonon focusing at the (100) face of silicon with a thin oxide overlayer. The oxide layer is assumed to be isotropic elastically and the focusing factors of both the Rayleigh-surface and pseudosurface phonons are calculated for several values of k (wave number) $\times$ h (thickness of the oxide layer). As kh increases we find that the strong focusing region of Rayleigh-surface phonons in between two caustic directions becomes narrow and moves toward the [110] direction. Also the directions around the [110] axis, where the pseudosurface phonons are allowed to exist, are diminished and eventually their spatial intensity distribution disappears. [S0163-1829(97)02308-4]

Ballistic phonon propagation in a crystalline solid is affected profoundly by the presence of the elastic anisotropy of the lattice, leading to the spatial focusing and defocusing of acoustic energy. This effect was first found for bulk phonons with a heat pulse method[1] and then studied in detail with phonon imaging techniques[2] in the 1980s. The ballistic phonon focusing at crystal surfaces was also predicted theoretically many years ago,[3,4] but it is quite recently that the effect was observed experimentally. An optical excitation of low-frequency, incoherent surface phonons in the 50-MHz range[5,6] and also an excitation of coherent surface acoustic waves of $\sim$ 15 MHz with focused immersion transducers[7,8] are employed for the observation of this effect. More recently, Höss and Kinder have tried an imaging experiment with a traditional heat pulse method.[9] They have used a (100) silicon surface for which the laser-annealing technique was applied to obtain as clean a surface as possible. Unfortunately, the signal of observed ballistic surface phonons is rather weak due to strong scatterings from surface roughness and an improved spatial resolution of surface-phonon images is necessary for a detailed comparison with existing theory. They also suggest the existence of a thin oxide layer (SiO_2) on the sample surface, which should affect the focusing of high-frequency surface phonons if phonon wavelengths are comparable to the thickness of the layer. The purpose of the present paper is to study theoretically the surface phonon focusing in the presence of a noncrystalline oxide overlayer considered to be elastically isotropic.

We note that the dominant phonon frequencies involved in a heat pulse (several hundred gigahertz) are in the region where the continuum elasticity theory is well applicable. The elastic-wave problem in a layered half space has already been studied extensively and the existence of several branches of surface waves has been found.[10] In the present study, however, we concentrate our discussion on the generalized Rayleigh phonons, which are reduced to the Rayleigh-surface phonons on a free-flat surface and also pseudosurface phonons. By the pseudosurface phonons we mean the acoustic mode for which the displacement of one of three partial waves in a silicon substrate grows exponentially with the distance from a Si-SiO_2 interface.[11]

We take a Cartesian coordinate system such that a Si substrate (a SiO_2 layer) occupies $x_3=z>0$ $(0>z>-h)$ and the surface of SiO_2 layer $(z=-h)$ and the Si-SiO_2 interface $(z=0)$ are parallel to the plane containing the vector $\mathbf{x}_\parallel=(x_1,x_2)=(x,y)$. Thus, in Si the displacement vector $\mathbf{u}$ takes the form

$$\mathbf{u}=\sum_{\alpha=1}^{3} A^{(\alpha)}\boldsymbol{\varepsilon}^{(\alpha)}\exp[i(\mathbf{k}_\parallel\cdot\mathbf{x}_\parallel+k_3^{(\alpha)}z-\omega t)], \qquad (1)$$

where $\mathbf{k}_\parallel=k_\parallel\mathbf{n}=(k_1,k_2)$ is the two-dimensional wave vector common to that in the oxide layer ($\mathbf{n}$ is the direction cosine of the wave vector, i.e., $|\mathbf{n}|=1$), ω is the angular frequency, α (=1, 2, and 3) discriminates three partial waves with amplitudes $A^{(\alpha)}$, $\boldsymbol{\varepsilon}^{(\alpha)}$'s are the unit polarization vectors, and $k_3^{(\alpha)}$'s determine the spatial variation of the lattice displacement perpendicular to the interface. The wave numbers $k_3^{(\alpha)}$'s are determined by solving the equation of motion with $\mathbf{k}=(\mathbf{k}_\parallel,k_3)$,

$$(\rho_S\omega^2\delta_{im}-C_{ijmn}k_jk_n)\varepsilon_m=0 \quad (i=1,2,3) \qquad (2)$$

for given $\mathbf{k}_\parallel$ and ω, where ρ_S is the mass density of silicon and C_{ijmn} is the stiffness tensor. The summation convention over repeated indices is assumed in Eq. (2). It should be noted that $\mathrm{Im}[k_3^{(\alpha)}]>0$ ($\alpha=1$, 2, and 3) for Rayleigh-surface phonons, but for pseudosurface phonons $\mathrm{Im}[k_3^{(\alpha)}]>0$ ($\alpha=1,2$) and $\mathrm{Im}[k_3^{(3)}]<0$. For the latter case the wave amplitude should decay as the wave propagates along the interface, so $\mathbf{k}_\parallel$ is a complex vector with a positive imaginary part.

Similarly, the displacement vector associated with the acoustic wave in the oxide layer is written as

$$\mathbf{U}=\sum_{\alpha=1}^{6} B^{(\alpha)}\mathbf{e}^{(\alpha)}\exp[i(\mathbf{k}_\parallel\cdot\mathbf{x}_\parallel+q^{(\alpha)}z-\omega t)], \qquad (3)$$

where $\mathbf{e}^{(\alpha)}$'s are the unit polarization vectors and $q^{(\alpha)}$'s determine the spatial variation of the partial waves inside the oxide layer perpendicular to the surface. Here we note that inside the layer the displacement vector consists of six partial waves that are indexed by α $(=1-6)$ with amplitudes $B^{(\alpha)}$ [three of them ($\alpha=1,2,3$) are $+z$ going and the other three ($\alpha=4,5,6$) are $-z$ going waves]. The wave numbers $q^{(\alpha)}$ ($q^{(\alpha+3)}=-q^{(\alpha)},\alpha=1,2,3$) satisfy the dispersion relations

$$k_{\|}^2+[q^{(1)}]^2=\omega^2/c_L^2\equiv K_L^2, \tag{4}$$

$$k_{\|}^2+[q^{(\alpha)}]^2=\omega^2/c_T^2\equiv K_T^2 \quad (\alpha=2,3), \tag{5}$$

where $\lambda+2\mu=\rho_0 c_L^2$ $\mu=\rho_0 c_T^2$ (ρ_0 is the density of the oxide layer and λ and μ are the Lame constants), and K_L and K_T are the magnitudes of the wave vectors associated with the longitudinal and transverse waves (with phase velocities c_L and c_T, respectively) in the layer. Explicitly, the polarization vectors are written as

$$\mathbf{e}^{(\alpha)}=(\mathbf{k}_{\|},q^{(\alpha)})/K_L \quad (\alpha=1,4), \tag{6}$$

$$\mathbf{e}^{(\alpha)}=(q^{(\alpha)}\mathbf{k}_{\|},-k_{\|}^2)/k_{\|}K_T \quad (\alpha=2,5), \tag{7}$$

$$\mathbf{e}^{(\alpha)}=(\mathbf{k}_{\perp},0)/K_T \quad (\alpha=3,6), \tag{8}$$

where $\mathbf{k}_{\perp}=(-k_2,k_1)$.

The stress $\boldsymbol{\sigma}$ induced at the (001) plane of silicon is given by

$$\sigma_j=iC_{44}\sum_{\alpha=1}^{3}A^{(\alpha)}(k_j\varepsilon_3^{(\alpha)}+k_3^{(\alpha)}\varepsilon_j^{(\alpha)})\times\exp[i(\mathbf{k}_{\|}\cdot\mathbf{x}_{\|}+k_3^{(\alpha)}z-\omega t)] \quad (j=1,2),$$

$$\sigma_3=i\sum_{\alpha=1}^{3}A^{(\alpha)}[C_{12}(k_1\varepsilon_1^{(\alpha)}+k_2\varepsilon_2^{(\alpha)})+C_{11}k_3^{(\alpha)}\varepsilon_3^{(\alpha)}]\times\exp[i(\mathbf{k}_{\|}\cdot\mathbf{x}_{\|}+k_3^{(\alpha)}z-\omega t)], \tag{9}$$

where C_{ij} are the elastic constants. Also the stress $\boldsymbol{\Sigma}$ at the plane normal to z axis in the oxide layer is given by

$$\Sigma_j=i\mu\sum_{\alpha=1}^{6}B^{(\alpha)}(k_je_3^{(\alpha)}+q^{(\alpha)}e_j^{(\alpha)})\times\exp[i(\mathbf{k}_{\|}\cdot\mathbf{x}_{\|}+q^{(\alpha)}z-\omega t)] \quad (j=1,2),$$

$$\Sigma_3=i\sum_{\alpha=1}^{6}B^{(\alpha)}[\lambda(k_1e_1^{(\alpha)}+k_2e_2^{(\alpha)})+(\lambda+2\mu)q^{(\alpha)}e_3^{(\alpha)}]\times\exp[i(\mathbf{k}_{\|}\cdot\mathbf{x}_{\|}+q^{(\alpha)}z-\omega t)]. \tag{10}$$

The surface wave velocities in the present system are determined from the stress-free boundary condition at the surface $z=-h$, i.e.,

$$\boldsymbol{\Sigma}(z=-h)=0, \tag{11}$$

and the continuity of both the lattice displacement and the stress at the interface $z=0$

$$\mathbf{u}(z=0)=\mathbf{U}(z=0), \tag{12}$$

$$\boldsymbol{\sigma}(z=0)=\boldsymbol{\Sigma}(z=0). \tag{13}$$

We have searched numerically for the surface wave velocities $c=\omega/k_{\|}$ that give zeros of the determinant of a 9×9 matrix derived from Eqs. (11)–(13).

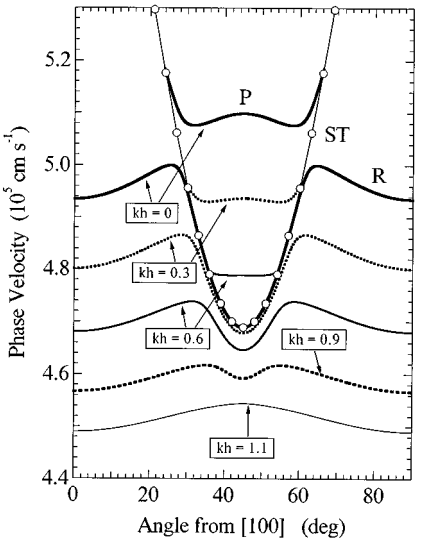

FIG. 1. Phase velocities of Rayleigh-surface phonons (R) and pseudosurface phonons (P) in the (001) plane of silicon with an oxide overlayer of normalized thickness kh. The velocities of the pseudosurface phonons appear in the upper region of the bulk slow transverse (ST) branch in silicon plotted by open circles connected by a thin solid line. The pseudosurface phonons do not exist for $kh>0.81$.

In Fig. 1 the calculated angular dependences of the phase velocities of both the Rayleigh-surface and pseudosurface phonons are plotted for five selected values of the normalized thickness $kh=\omega h/v_T^{[100]}$ of the layers, where $v_T^{[100]}$ is the sound velocity of the bulk slow-transverse (ST) phonons propagating in the [100] direction of silicon.[12] The boldest solid lines show the results for the free (100) surface of silicon ($kh=0$). The velocity of the Rayleigh-surface phonons on the free surface of SiO_2 is 3.35 km/s, which is much slower than the velocities plotted in this figure.

As expected, the angular dependence of the phase velocities becomes small as the layer thickness increases because the system becomes more like an isotropic continuum consisting of only SiO_2. For a finite layer thickness the pseudosurface phonons that do not exist in isotropic solid surfaces are allowed only in a small region near the [110] direction (45°). An interesting observation is that the pseudosurface phonons always appear in the upper part of the bulk ST branch in the (100) plane of the Si substrate (open circles connected by a thin solid line). This means that the velocities of the pseudosurface phonons in the present system are necessarily larger than those of the bulk ST phonons propagating in the same plane of the silicon substrate. This is due to the fact that the decay of the wave amplitude characteristic of the pseudosurface branch is caused only when the surface phonons have phase velocities faster than the slowest bulk phonons and hence the pseudosurface phonons are unstable against the coupling with this bulk branch.[11,13]

For a value of kh larger than 0.9 for which the velocities of the Rayleigh-surface phonon are located well below the ST branch, the pseudosurface phonons do not appear any more. More precisely, for $kh=0.81$ a very small pseudosurface branch appears at the bottom of the ST branch, but for $kh=0.82$ we could not find any reliable solutions for the pseudobranch.

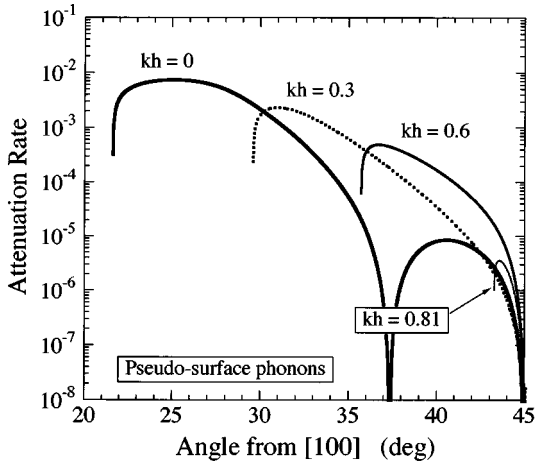

FIG. 2. Angular dependence of the attenuation rate of pseudosurface phonons in the (001) plane of silicon with an oxide overlayer. No solution was found for $kh \geq 0.82$.

The attenuation rate of the pseudosurface phonons parallel to the surface defined by $\mathrm{Im}[k_{\parallel}]/\mathrm{Re}[k_{\parallel}]$ is plotted in Fig. 2 for four chosen values of kh (as noted above, $kh=0.81$ is the upper limit for which the solution of the pseudobranch is found). As kh increases the maximum value of the attenuation rate decreases and these phonons behave more like true surface phonons. The origin of these small attenuations along the surface is the weak coupling between the pseudosurface phonons polarized predominantly in the saggital plane and the bulk ST phonons that are polarized almost perpendicular to the same plane.[11,13] It should be noted that on the free surface of silicon ($kh=0$) the phonons in the pseudo-surface branch become true surface phonons at an angle $\sim 37.3°$. This happens only at small values of kh (<0.2).

The focusing factors are plotted in Fig. 3. The idea for quantifying the surface phonon focusing is much the same as that employed for the bulk phonons.[1] We define the focusing factor A by $A=|\Delta\theta_{\mathbf{k}}/\Delta\theta_{\mathbf{v}}|$, where $\Delta\theta_{\mathbf{k}}$ is a small angle occupied by the wave vector $\mathbf{k}_{\parallel}$ in the wave-vector space ($\mathbf{k}_{\parallel}$ space) and $\Delta\theta_{\mathbf{v}}$ is the corresponding small angle subtended by the group-velocity vector $\mathbf{v}$ in the real space. In the calculation of A we assume a uniform distribution for the wave-vector directions in the $\mathbf{k}_{\parallel}$ space. Thus, the focusing factor A plotted for $\theta_{\mathbf{v}}$ gives the enhancement of the surface phonon flux for a specified direction in the real space ($A=1$ in isotropic solids). Geometrically this number is inversely proportional to the curvature of the slowness curve (defined by $|\mathbf{k}_{\parallel}|=\omega/c$ with a constant ω) of surface phonons in the $\mathbf{k}_{\parallel}$ space.[14] Hence, A diverges in the directions corresponding to the inflection points of the slowness curve, which separate the concave regions from the convex regions. Such directions in the real space are called caustics.

As noted above, the variation of sound velocities with respect to propagation direction becomes small as the layer thickness h increases. This leads to the fact that the concave region of the slowness curve of the Rayleigh-surface phonons,[15] which induces the strong focusing effect, shrinks and the slowness curve becomes more like a circle. Thus the narrowing of the Rayleigh-phonon focusing region happens, though caustics continue to exist up to $kh \simeq 0.9$. At the same time the strong focusing region in between nearby caustics

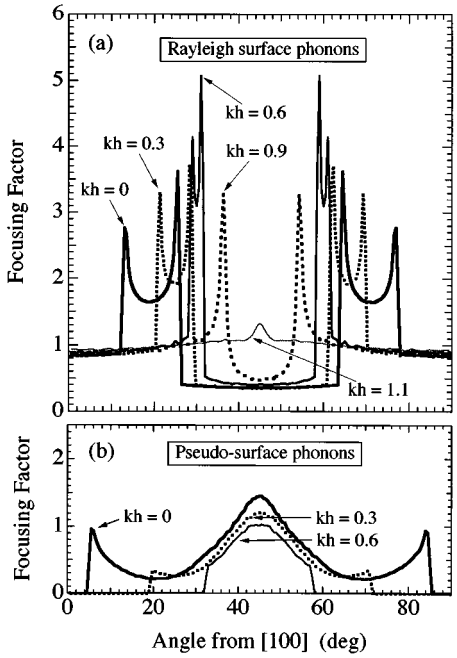

FIG. 3. Angular dependences of the focusing factors of (a) Rayleigh-surface phonons and (b) pseudosurface phonons in the (001) plane of silicon with an oxide overlayer of normalized thickness kh.

moves toward the [110] axis. For a value of $kh \sim 0.9$ the concave region on the slowness curve vanishes and a pair of caustics coalesce. After the associated coalescence of two sharp enhancements in the focusing factor (at caustics) only bumps in enhancement of phonon flux are produced around the [110] axis and its equivalent directions. As the layer thickness increases we also see that the rather moderate angular-dependent focusing in the pseudobranch on either side of the [110] direction becomes weaker (no caustic exists) and the region of allowed energy propagation is restricted closer to the [110] axis. For values of kh larger than 0.82 the intensity distribution of the pseudosurface phonons disappears.

To summarize, we have studied the effects of isotropic oxide layer on the focusing of high-frequency Rayleigh-surface and pseudosurface phonons propagating on the (100) face of silicon. As the layer thickness increases the waves localized near the surface of the system become more like the surface waves in an isotropic elastic medium. Therefore, the angular dependence of Rayleigh-surface phonon velocity becomes small and the allowed directions of pseudosurface phonons characteristic of an anisotropic substrate are diminished. This means that the focusing region of Rayleigh-surface phonons shrinks with increasing the layer thickness but an important result is the fact that the caustics also move appreciably toward the [110] axis. The experimental surface phonon images obtained by Höss and Kinder[9] exhibit a broad enhancement in phonon intensity that covers the theoretical focusing region of Rayleigh-surface phonons on the free (100) silicon surface. However, no caustics predicted are resolved. So, at present, we cannot say anything about possible existence of an oxide layer on the silicon surface. We hope further imaging experiments on the high-frequency surface phonon propagation are executed with improved spatial reso-

lutions. Also the present work suggests that the measurement of caustic directions should provide us with a possible tool for estimating the thickness of an oxide layer if the dominant frequency of surface phonons excited is known.

Finally, we note that the above results obtained are not restricted to the (100) silicon, but should be valid qualitatively for the focusing of surface phonons on the (100) face of other cubic crystals with soft overlayers. Such crystals are Ge and GaAs, which possess the anisotropy ratio $(C_{11}-C_{12})/2C_{44}$ smaller than unity.

S. T. wishes to thank C. Höss and H. Kinder for valuable discussions. This work was supported in part by the Murata Science Foundation.

[1] B. Taylor and H. J. Maris, Phys. Rev. Lett. **23**, 416 (1969); H. J. Maris, J. Acoust. Soc. Am. **50**, 812 (1971).

[2] G. A. Northrop and J. P. Wolfe, in *Nonequilibrium Phonon Dynamics*, edited by W. E. Bron (Plenum, New York, 1985), Chap. 5.

[3] S. Tamura and K. Honjo, Jpn. J. Appl. Phys. Suppl. 3 **20**, 17 (1980); K. Honjo and S. Tamura (unpublished).

[4] R. E. Camley and A. A. Maradudin, Phys. Rev. B **27**, 1959 (1983).

[5] Al. A. Kolomenskii and A. A. Maznev, JETP Lett. **53**, 423 (1991); Phys. Rev. B **48**, 14 502 (1993).

[6] A. A. Maznev, Al. A. Kolomenskii, and P. Hess, Phys. Rev. Lett. **75**, 3332 (1995); A. A. Maznev and A. G. Every, Solid State Commun. **97**, 1679 (1996).

[7] R. E. Vines, S. Tamura, and J. P. Wolfe, Phys. Rev. Lett. **74**, 2729 (1995); **75**, 1873(E) (1995).

[8] R. E. Vines, M. R. Hauser, and J. P. Wolfe, Z. Phys. B **98**, 255 (1995).

[9] C. Höss and H. Kinder, Physica B **219&220**, 706 (1996).

[10] G. W. Farnell and E. L. Adler, in *Physical Acoustics XI*, edited by W. P. Mason and R. N. Thurston (Academic, New York, 1972), p. 35.

[11] T. C. Lim and G. W. Farnell, J. Acoust. Soc. Am. **45**, 845 (1969).

[12] We used the elastic constants $C_{11}=16.76$, $C_{12}=6.51$, and $C_{44}=8.01$ (in units of 10^{11} dyn/cm^2) and the mass density $\rho_S=2.33$ g/cm^3 for silicon, and the mass density $\rho_0=2.20$ g/cm^3 and $c_L=5835$ m/s and $c_T=3711$ m/s for the longitudinal and transverse sound velocities in SiO_2. The sound velocity $v_T^{[100]}$ in silicon is 5863 m/s.

[13] S. Tamura, R. E. Vines, and J. P. Wolfe, Phys. Rev. B **54**, 5151 (1996).

[14] H. Shirasaki and T. Makimoto, J. Appl. Phys. **50**, 2795 (1979).

[15] For the shapes of the slowness curves of the Rayleigh-surface and pseudosurface phonons at the (100) silicon ($h=0$), see S. Tamura and M. Yagi, Phys. Rev. B **49**, 17 378 (1994).