Diffuse scattering of high-frequency phonons at solid surfaces

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The effects of the mode conversion of high-frequency phonons on the diffuse scattering are discussed. The shape of phonon-reflection signals is obtained as a function of time for transverse phonons. The conclusion is that the mode-converted surface phonons are of considerable importance in order to interpret the cause of diffuse scattering of high-frequency phonons.

I. INTRODUCTION

The scattering of high-frequency (GHz to THz) phonons at crystal surfaces has attracted a good deal of experimental attention over the last few years.[1] Taborek and Goodstein[2] found in sapphire surfaces that there exist both specular and diffuse signals in their high-resolution phonon-reflection time-of-flight spectra. In addition, when liquid ^{4}He is present, the diffuse signals were severely affected. Subsequent studies on diffuse scattering of high-frequency phonons by heat pulse,[3] phonon imaging,[4–6] and thermal conduction[7,8] have improved our knowledge on the boundary scattering. The underlying physical basis of diffuse scattering is, however, still not well understood.

This paper aims to investigate the cause of diffuse scattering of high-frequency phonons. It will be shown analytically that diffuse signals are due to two causes. One comes from the direct scattering at irregular surface, bulk phonons$\rightarrow$bulk phonons (hereafter referred to as B phonon), and the other due to the process B phonons$\rightarrow$surface phonons (referred to as R phonon), where the mode-converted R phonons are rescattered back into B phonons by surface irregularities and constitute the diffuse signals. The scattering of transverse phonons propagating in a sapphire crystal is illustrated. Sapphire is a mild anisotropic crystal which allows one to use the isotropic elastic approximation, if the phonon focusing effect is disregarded.

II. FORMULATION OF DIFFERENTIAL CROSS SECTION OF HIGH-FREQUENCY PHONONS

The displacement vector in an isotropic solid with a free boundary can be expanded in terms of eigenmodes:[9]

$$\mathbf{u}(\mathbf{x},t)=\sum_J \left[\frac{\hbar}{2\rho\omega_J S}\right]^{1/2}[a_J \mathbf{u}_J(z)e^{i\mathbf{k}\cdot\mathbf{r}-i\omega t}+\text{H.c.}] , \qquad (1)$$

where ρ is the mass density of the medium and $J=(\mathbf{k},c,m)$ labels a set of quantum numbers which specifies the *eigenmodes* of phonons, where $\mathbf{k}$ is a two-dimensional wave vector, c the velocity of a *wave front* traversing the surface, and m specifies the mode. The sum over J in Eq. (1) is defined as

$$\sum_J=\frac{S}{(2\pi)^2}\int d^2k\left[f(\mathbf{k}_R,c_R,R)+\sum_{m\,(\neq R)}\int_{D_m}\frac{dc}{c}f(\mathbf{k},c,m)\right] , \qquad (2)$$

where D_m denotes the spectral range of the velocity c. In Eq. (1), a_J and $a_J^\dagger$ are annihilation and creation operators, respectively, of the J-mode phonons.

There exist two different kinds of transverse modes. The first mode is the transverse mode with parallel polarization to a surface (referred to as the $T_\parallel$ mode), in which the angle of incidence $\theta_\parallel$ (defined from the vertical axis to a surface) is related to the velocity c through the definition

$$\cot\theta_\parallel=\beta(c)=[(c/c_{\rm ST})^2-1]^{1/2}$$

and the spectral range of c is from $c_{\rm ST}$ (corresponding to $\theta_\parallel=\pi/2$) to infinity ($\theta_\parallel=0$). In the present work the velocity of this mode is identified as $c_{\rm ST}$ indicating the slow-transverse waves. Transverse phonons polarized in the sagittal plane ($T_\perp$ phonons) are divided into two orthogonal modes, in which one has the evanescent pseudo-surface-wave ($c_{\rm FT}\le c\le c_{\rm L}$) and the others consist of longitudinal and transverse bulk waves interacting with each other through the surface ($c\ge c_{\rm L}$). Here $c_{\rm FT}$ and $c_{\rm L}$ mean the velocities of the fast-transverse and longitudinal modes, respectively. The explicit forms of these waves are given in Ref. 9.

The scale of roughness of sapphire is known to be of the order of 100 Å as described in Refs. 2 and 4–6. The surface roughness can be described as the mass defect localized in the vicinity of the surface.[10] The decay rate (i.e., the reciprocal of lifetime) of the J-mode phonon due to mass defect is given by the following formula:[11]

$$\Gamma_J=\sum_{J'}\left[\frac{\pi}{2\rho^2 S}\right]\delta(\omega_J-\omega'_J)\omega_{J'}^2 \times\left|\int_0^\infty dz\,\mathbf{u}_J(z)\cdot\mathbf{u}_{J'}(z)D(z)\right|^2\langle\,|\Delta\rho(\mathbf{k}+\mathbf{k}')|^2\rangle , \qquad (3)$$

where $D(z)$ represents the distribution of the density fluctuation in the vicinity of solid surfaces. The Fourier transform of the density-correlation function is defined by

$$\langle\,|\Delta\rho(\mathbf{k}+\mathbf{k}')|^2\rangle=\int d\mathbf{r}\,e^{i(\mathbf{k}+\mathbf{k}')\cdot\mathbf{r}}\langle\Delta\rho(\mathbf{r})\Delta\rho(\mathbf{0})\rangle\ .$$

The part of mass-density fluctuation $\Delta\rho(\mathbf{r})$ comes from the separation of the average mass density [i.e., $\rho(\mathbf{x})=\rho_0+\Delta\rho(\mathbf{r})D(z)$]. The angular brackets denote an ensemble average on a random function $\Delta\rho(\mathbf{r})$. In order to clarify the physical origin of the problem in a simple fashion, the surface roughness is treated as a white noise and is assumed to be localized in the vicinity of the surface [i.e., $D(z)=\delta(z)$]. These are valid for phonons with longer wavelength compared with the roughness scale ($\lambda>100$ Å in the present work). Hence, the ensemble average is obtained as

$$\langle\,|\Delta\rho(\mathbf{k}+\mathbf{k}')|^2\rangle=(\overline{\Delta\rho})^2a^2\ ,$$

where a is a characteristic length of roughness.

First, let us consider the case where $T_{\parallel}$ phonons are incident at angle $\theta_{\parallel}$ on a rough surface and are scattered into B phonons and R phonons. After straightforward calculation using the wave functions given in Ref. 9 and the definition of Eq. (2), the differential cross section for the process $T_{\parallel}+\text{roughness}\rightarrow T'_{\parallel}$ becomes

$$d\sigma(T_{\parallel}\rightarrow T'_{\parallel})=\frac{A\cos^2\varphi}{c_{\rm ST}^4x^2(x^2-1)^{1/2}}d\varphi\,dx\ ,\quad x=c/c_{\rm ST} \tag{4a}$$

where

$$A=\frac{(\overline{\Delta\rho}\,a)^2\omega^4}{8\pi^2\rho^2}\ . \tag{4b}$$

Here the cross section was obtained from $d\sigma(J\rightarrow J')=\hbar\omega_J\Gamma_{J\rightarrow J'}/(\text{energy flux})$. φ is the azimuthal angle and x is related to the polar angle $\theta_{\parallel}$ through the relation $\cot\theta_{\parallel}=(x^2-1)^{1/2}$. The cross sections into $T_{\perp}$ phonons with the spectral range of c larger than $c_{\rm L}$ and $c_{\rm FT}\le c\le c_{\rm L}$ are obtained as

$$d\sigma(T_{\parallel}\rightarrow T'_{\perp})=\frac{A\sin^2\varphi\beta'\,|\,\zeta'_{\pm}\,|^2d\varphi\,dy}{c_{\rm ST}c_{\rm L}^3y^4}\ ,\quad y=c/c_{\rm L} \tag{5}$$

and

$$d\sigma(T_{\parallel}\rightarrow T''_{\perp})=\frac{A\sin^2\varphi\beta'\,|\,A'\,|^2d\varphi\,dx}{c_{\rm ST}c_{\rm FT}^3x^4}\ . \tag{6}$$

For the process $T_{\parallel}+\text{roughness}\rightarrow R$, the cross section becomes

$$d\sigma(T_{\parallel}\rightarrow R)=\frac{\pi A\sin^2\varphi f_1^2}{2c_{\rm ST}c_R^3K}d\varphi\ , \tag{7}$$

and the numerical factors f_1 and K are written as

$$f_1=1-2\gamma\eta/(1+\eta^2)\ ,$$

$$K=(\gamma-\eta)(\gamma-\eta+2\gamma\eta^2)/2\gamma\eta^2\ .$$

The factors $(A,\beta,\gamma,\eta,\zeta)$ appearing in Eqs. (5)—(7) are defined in the Appendix of Ref. 11. The differential cross section $d\sigma(T_{\perp}\rightarrow J')$ for $T_{\perp}$ phonons can be obtained by a similar manner whose expressions are omitted here.

III. CALCULATED RESULTS

A. Diffuse signals from directly scattered *B* phonons

When transverse (T) phonons are emitted according to the Lambertian law from the heater attached to the crystal of thickness of h, then each element of the area $dA=r\,dr\,d\varphi$ on the top surface is irradiated by phonons from the heater (see the inset in Fig. 1). If the heater and bolometer are assumed to be very small and close together, the bolometer detects only the phonons backscattered with the same angle as that of the incident phonons, i.e., the element dA can be considered as a new source.[2] We have the fraction of the reflected intensity by defining the heat flux as $S(t)$ emitted by the heater:

$$dR_1=\sum_{J'(\neq R)}d\sigma^*(x,T\rightarrow J')\frac{\cos^2\theta S(t-2l/c_{J'})}{l^4}dx\,d\varphi\ , \tag{8}$$

where $l^2=h^2+r^2$, and the "cross section"

$$d\sigma^*(T\rightarrow J')\equiv d\sigma(T\rightarrow J')/dx\,d\varphi\ .$$

The decay into R phonons from T phonons is not included here; those effects will be discussed later. The diffuse signal R_1 as a function of time is obtained by integrating Eq. (8) over φ and x by taking the heat pulse given by a δ function $S(t)=S_0\delta(t)$. This assumption is good for the crystals of the order of 1 cm in height because heat pulses used in the experiments were of 10—100 nsec duration.[2] The relation between r in Eq. (8) and $x=c/c_{\rm ST}$ is obtained by using $\cot\theta_{\parallel}=(x^2-1)^{1/2}$ where $x^2=(h^2+r^2)/r^2$. From Eq. (8), the diffuse signal is represented by

$$R_1(t)=S_0\sum_{J'\neq R}\int_0^{2\pi}d\varphi\int_{D_m}dx\frac{d\sigma^*_{J'}\cos^2\theta\delta(t-2l/c_{J'})}{(r^2+h^2)^2}\ . \tag{9}$$

Finally the integration of Eq. (9) gives, by defining $R_1=\sum_i r_i$,

$$r_1(T_{\parallel}\rightarrow T'_{\parallel})=\frac{C_1}{T_1^8}\ ,\quad T_1\ge h \tag{10}$$

$$r_2(T_{\parallel}\rightarrow T'_{\perp})=\frac{C_2}{T_2^{10}}\ ,\quad T_2\ge h/d_1 \tag{11a}$$

$$r_3(T_{\perp}\rightarrow T'_{\perp})=\frac{C_3}{T_2^{12}}[h^4+2(T_3^2-h^2)^2]\ ,\quad T_3\ge h/d_2\ . \tag{11b}$$

Here C_i are given as $C_1=\pi AS_0h^3/2c_{\rm ST}^3$, $C_2=\pi AS_0h^5/2\bar{c}^3$, $C_3=\pi AS_0h^3/2c_{\rm FT}^3$, $\bar{c}=2c_{\rm FT}c_{\rm ST}/(c_{\rm ST}+c_{\rm FT})$ and the definitions are $d_1=\bar{c}/c_{\rm ST}$, $d_2=c_{\rm FT}/c_{\rm ST}$; $T_1=c_{\rm ST}t/2$, $T_2=\bar{c}t/2$, and $T_3=c_{\rm FT}t/2$.

B. Role of *R* phonons on the diffuse component

To see the effect of mode-converted R phonons on the diffuse signals, we consider first the decay rate of R phonons by roughness (note that the mode-converted B phonons at rough surfaces are assumed to reach the detector without scattering in the present system). The decay rate of R phonons into T phonons can be obtained by using Eq. (3) as well,

$$\frac{1}{\tau_R} \simeq \frac{(\overline{\Delta\rho}\, a)^2 \omega^5 f_1^2}{4\pi\rho^2 c_R c_{\mathrm{ST}}^3 K} . \tag{12}$$

The sapphire surfaces used in experiments[2,4–6] have a roughness scale of the order of $\delta = 100$ Å. This indicates that one can treat the surface with a mean variation in depth and width of $\delta = 100$ Å with the areal density $w = 0.5\delta^{-2}$.[10] The characteristic length δ of roughness should correspond to the length scale a in Eq. (4b) and one can replace $(\overline{\Delta\rho}\, a)^2$ in the cross section by $w(\Delta M)^2$, where the average mass of "bumps" is $\Delta M = \rho a^3$. The use of the known values for sapphire, $c_{\mathrm{L}} = 11\times 10^5$ cm/sec, $c_{\mathrm{ST}} = 6\times 10^5$ cm/sec, $c_R = 0.92 c_{\mathrm{ST}}$ and $\rho = 3.99$ g/cm^3, in Eq. (12) gives

$$\tau_R \sim 100\nu^{-5} \text{ sec} , \tag{13}$$

where ν is in the GHz range. By taking $\nu = 50$—100 Ghz, we have $\tau_R \simeq 10$ nsec. It is too small to distinguish this time-delay effect in the diffuse tail within experimental accuracy (since the time duration of heat pulse is 10—100 nsec). Thus we can neglect the time-delay effect in the analysis of the diffuse tail for the process of mode-converted R phonons.

The ratio of intensities between the two processes $\mathrm{T}_{\parallel} \rightarrow J'$ and $\mathrm{T}_{\parallel} \rightarrow R \rightarrow J''$ becomes

$$\gamma = \frac{d\sigma(\mathrm{T}_{\parallel} \rightarrow R' \rightarrow J')}{d\sigma(\mathrm{T}_{\parallel} \rightarrow J')} = \frac{\pi f_1^2}{2K} \left[\frac{c_{\mathrm{ST}}}{c_R} \right]^3 \simeq 0.2 . \tag{14}$$

This ratio does not depend on the reflected angle $\theta_{\parallel}$, which implies that the ratio of intensities is the same at any time t in the diffuse tail.

The calculated result of T-phonon reflection signals as a function of time is shown in Fig. 1 for the direct process (A) and the process (B) from the mode-converted R phonons. The parameters for sapphire are used: $h = 1$ cm, $c_{\mathrm{L}} = 11.0\times 10^5$ cm/sec, $c_{\mathrm{ST}} = 6.0\times 10^5$ cm/sec, and $c_{\mathrm{FT}} = 6.5\times 10^5$ cm/sec. Curve 2 at 3.2 μsec corresponds to the mode-converted $\mathrm{T}_{\parallel}$ phonons and $\mathrm{T}_{\perp}$ phonons. Curves 1 and 3 come from the processes $\mathrm{T}_{\parallel} \rightarrow \mathrm{T}'_{\parallel}$ and $\mathrm{T}_{\perp} \rightarrow \mathrm{T}'_{\perp}$, respectively.

IV. CONCLUSIONS

In summary, a quantitative study has been made of the diffuse scattering of high-frequency phonons from the sapphire surface. The results imply that the diffuse signals are composed of two causes: one comes from direct scattering (B phonons + roughness $\rightarrow B$ phonons) and the second is due to the process B phonons + roughness $\rightarrow R$ phonons $\rightarrow B$ phonons. The calculated results recover the experimental features well.[2,3] The mode-converted R phonons are of considerable importance in order to interpret the origin of the diffuse scattering of high-frequency phonons as seen from Fig. 1. The contribution from L phonons to the diffuse signals has not been discussed in the present paper. It is known[2,3,12] that the diffuse signals are severely affected by placing liquid He at the solid surface. In this connection, the mode-converted R phonons should play an important role for the energy transfer into liquid helium.[13] In addition, when the inelastic scattering sources are present in the irregular surface,[7] the mode-converted R phonons should be responsible for the effective energy absorption because R phonons possess a localized nature in the vicinity of the surface.

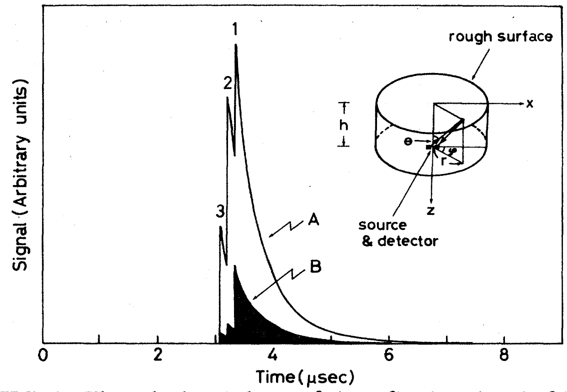

FIG. 1. The calculated shape of the reflection signal of high-frequency T phonons at the sapphire surface is shown as a function of time. Curve A comes from the mode conversion to B phonons. Curve B is from the component of the mode-converted R phonons. The thickness of the crystal is taken to be $h = 1$ cm. The largest peak (1) at $t = 2h/c_{\mathrm{ST}} \simeq 3.31$ μsec corresponds to the process $\mathrm{T}_{\parallel} \rightarrow \mathrm{T}'_{\parallel}$. The middle peak (2) is due to the process $\mathrm{T}_{\parallel} \rightarrow \mathrm{T}'_{\perp}$ and $\mathrm{T}_{\perp} \rightarrow \mathrm{T}'_{\parallel}$. The small peak (3) is attributed to the process $\mathrm{T}_{\perp} \rightarrow \mathrm{T}'_{\perp}$. Note that the ratio of heights of the largest peaks of curves A and B is 0.2. The inset shows the geometrical arrangement of the present system.

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