

## Resonant interaction of phonons with surface vibrational modes in a finite-size superlattice

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We study theoretically the interaction of phonons with surface vibrational modes in a finite-size superlattice with a free surface. A phonon incident normally on the superlattice from a substrate is perfectly reflected, i.e., the reflection rate is unity irrespective of frequency. However, it comes back to the substrate with a large time delay when the frequency coincides with an eigenfrequency of the surface mode. This result is attributable to the resonant interaction of incident phonons with a vibrational mode localized near the surface.

Acoustic vibrations in superlattices (SL's) with various stacking order, such as periodic, quasiperiodic, and random superlattices have been investigated extensively during the last decade.<sup>1-11</sup> The SL's actually grown are not ideal and they usually possess both natural and artificial defects. In particular, an inhomogeneity embedded in a SL with perfect periodicity (e.g., a defect layer or a free surface) is shown to cause localized vibrations within the frequency gaps induced by the periodicity of a SL. The localized vibrational modes due to defect layers have also been studied theoretically for both infinite- and finite-size SL systems. For an infinite, periodic SL with a defect layer, it is possible to derive analytically the frequencies of these localized modes.<sup>6</sup> For a finite-size system, on the other hand, the transmission and reflection rates have been calculated, and their local enhancements are indicative of the presence of the localized vibrational states.<sup>7</sup> However, the study of the surface vibrations or surface acoustic waves in SL's was, hitherto, carried out only for a semi-infinite system.<sup>8,9</sup>

In the present paper, we study the interaction of the surface vibrational modes with acoustic phonons injected to a finite-size superlattice grown on a substrate (see Fig. 1). Specifically, we consider a possibility of observing surface vibrations by a phonon reflection experiment. The phonons incident on a SL with a frequency inside a gap are Bragg reflected, i.e., the associated lattice displacement decays exponentially in the SL with the distance from the substrate. The phonons within a frequency band can propagate up to the surface of the SL but they are also perfectly reflected from the surface. In other words, the reflection rate in this system is unity for the whole frequency range. Thus, one may naively think that it is impossible to observe evidence of surface vibrations by a phonon reflection experiment. However, this is not the case. Although the magnitude of the reflection amplitude does not give us any information on the surface mode, its phase contains important information on the interaction with the surface vibrations, which is basically accessible in a phonon reflection experiment. This is because the time needed for a phonon to complete the reflection process is given by the derivative of this phase with respect to the frequency, or "phase time."<sup>12-14</sup> In particular, we show that for an incident phonon with a frequency near an eigenfrequency of surface vibrations the phase time is enhanced strongly due to the resonant interaction with the surface mode.

We can formulate the reflection of phonons in the present system as a stationary scattering problem in one dimension.<sup>10,11</sup> We adopt a continuum model and consider the phonon propagation normal to the layer interfaces. Thus, the displacement field of incident phonons is represented by the plane wave  $e^{ikx}$  ( $k$  is a wave number and  $x$  is normal to the layer interfaces), and that of reflected phonons is expressed as  $r(k)e^{-ikx}$  in the substrate region, where  $r(k)$  is the reflection coefficient. For a finite-size SL grown on a substrate and capped with a "detector layer," the analytical expression of the reflection coefficient is derived in a previous paper.<sup>10</sup> Setting the acoustic impedance of the detector layer to zero in Eq. (45) of Ref. 10, we can obtain the analytical form of  $r$  for a finite-size SL with a free surface. The result is

$$r = e^{i\Theta}, \tag{1}$$

where

$$\Theta = 2 \tan^{-1} \left( \frac{Z_X F}{Z_A \zeta} \right), \tag{2}$$

$$F = \frac{\mu - \lambda}{2} + \frac{C(N)}{S(N)}, \tag{3}$$

and

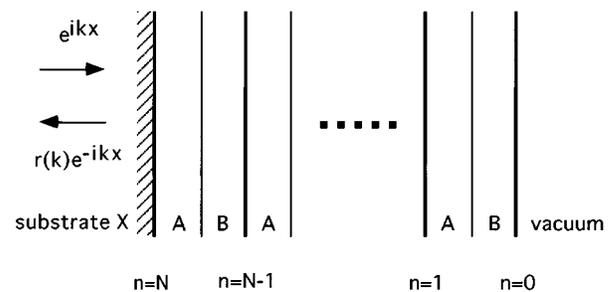


FIG. 1. Schematic of the system consisting of a substrate ( $X$ ) and a finite-size, periodic superlattice with alternate stacking of  $A$  and  $B$  layers.  $N$  denotes the number of bilayers and  $n$  indicates the boundaries of unit period. Phonons with a unit amplitude incident to the superlattice are reflected back to the substrate with reflected amplitude (reflection coefficient)  $r$ .

$$\zeta = -\sin k_A d_A \cos k_B d_B - \frac{Z_B}{Z_A} \cos k_A d_A \sin k_B d_B. \quad (4)$$

$$\mu = \cos k_A d_A \cos k_B d_B - \frac{Z_B}{Z_A} \sin k_A d_A \sin k_B d_B, \quad (5)$$

$$\lambda = \cos k_A d_A \cos k_B d_B - \frac{Z_A}{Z_B} \sin k_A d_A \sin k_B d_B. \quad (6)$$

In these equations,  $N$  is the number of the periodicity of a SL;  $d_A$  and  $d_B$  are the thicknesses of  $A$  (the layer adjacent to the substrate  $X$ ) and  $B$  layers, respectively;  $Z_i = \rho_i v_i$  ( $i=A, B$ , and  $X$ ) is the acoustic impedance given by the product of the mass density  $\rho_i$  and the sound velocity  $v_i$ ;  $k_i = \omega/v_i$  and  $\omega$  is the angular frequency. For phonons within a frequency gap (the frequency gap is determined by the condition  $|\mu + \lambda|/2 > 1$ ),  $S(N)$  and  $C(N)$  have the following forms:

$$S(N) = \left( \frac{\mu + \lambda}{|\mu + \lambda|} \right)^{N+1} \frac{\sinh N\theta}{\sinh \theta}, \quad (7)$$

$$C(N) = \left( \frac{\mu + \lambda}{|\mu + \lambda|} \right)^N \cosh N\theta, \quad (8)$$

and  $\theta = \cosh^{-1}(|\mu + \lambda|/2)$ . For a phonon within a frequency band ( $|\mu + \lambda|/2 \leq 1$ ), Eqs. (7) and (8) take different forms:  $S(N) = \sin N\theta/\sin \theta$ ,  $C(N) = \cos N\theta$ , and  $\theta = \cos^{-1}(|\mu + \lambda|/2)$ .

In both cases, the reflection rate  $R$  defined by  $|r|^2$  is unity. Thus, the reflection rate gives us no information on the interaction of phonons with the superlattice system. The relevant information is included in the phase  $\Theta$  of the reflection coefficient. This phase is related to the dynamical properties of phonons propagating through the system. Explicitly, the phase time  $\tau$  defined by  $d\Theta/d\omega$  describes the temporal delay of a reflected phonon packet. The phase time was originally introduced<sup>12–14</sup> for electrons tunneling through a potential barrier and later applied to the photon<sup>15</sup> and phonon propagations<sup>16,17</sup> in multilayered dielectric and elastic media. For recent reviews, see Refs. 18–21.

As an example, we show in Fig. 2(a) the frequency dependence of the phase time  $\tau$  calculated numerically for the  $(\text{AlAs})_{15}(\text{GaAs})_{15}$  superlattice with  $N=8$ . The plotted phase time oscillates around  $\tau_0 = 2N(d_A/v_A + d_B/v_B) = 0.0264$  ns (the time needed for a round trip) and exhibits sharp peaks in the lowest and third frequency gaps. These peaks, or large time delays associated with reflected phonons, are due to the resonant interaction with the localized surface vibrations which exist in these frequency gaps.

To understand this striking feature of the phase time, we derive an approximate expression of  $\tau$ . The function  $\zeta(\omega)$  defined in Eq. (4) can be rewritten as

$$\zeta = -\sin(\pi\omega/\omega_1) - \varepsilon \cos(\omega d_A/v_A) \sin(\omega d_B/v_B), \quad (9)$$

where  $\omega_1$  is the frequency at the center of the lowest frequency gap satisfying  $\cos\theta(\omega_1) = (\mu + \lambda)/2|_{\omega_1} = 1$ , or  $\omega_1 = \pi(d_A/v_A + d_B/v_B)^{-1}$  and  $\varepsilon = Z_B/Z_A - 1$  measures the acoustic mismatch between the constituent layers of the SL. For most of the SL's,  $Z_A$  is close to  $Z_B$ , i.e.,  $|\varepsilon| \ll 1$  ( $|\varepsilon| = 0.188$  for the longitudinal phonons in AlAs/GaAs

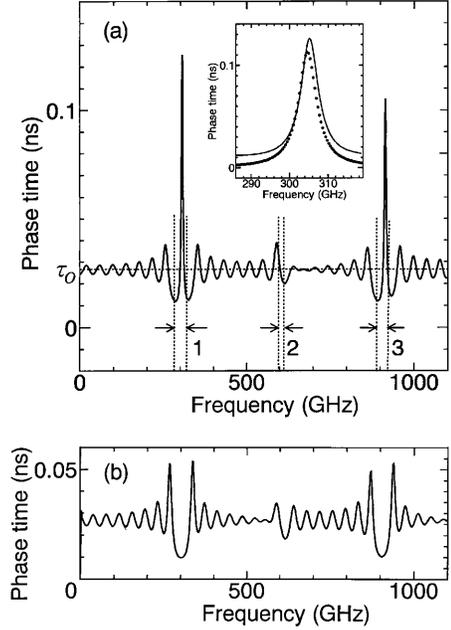


FIG. 2. Frequency dependence of the phase time of the longitudinal phonons reflected from a  $(100)\text{GaAs}/\text{AlAs}$  superlattice. The solid line and dots are the phase times calculated from the exact expression Eq. (2) and the approximate expression Eq. (15), respectively. Narrow frequency range labeled 1 to 3 indicates the lowest three frequency gaps of phonons. The unit period assumed in these calculations is  $(\text{GaAs})_{15}(\text{AlAs})_{15}$ : the surface layer is assumed to be (a) AlAs, i.e.,  $A=\text{GaAs}$  and  $B=\text{AlAs}$ , and (b) GaAs, i.e.,  $A=\text{AlAs}$  and  $B=\text{GaAs}$ . The parameters used are as follows: the number of periods is eight; the thickness of one monolayer is  $2.83 \text{ \AA}$  in the  $(100)$  direction for both GaAs and AlAs; the mass densities and longitudinal sound velocities are  $5.36 \text{ g/cm}^3$  and  $4.71 \text{ km/s}$  for GaAs, and  $3.76 \text{ g/cm}^3$  and  $5.65 \text{ km/s}$  for AlAs. In (a), the phase time within the lowest frequency gap is enlarged in the inset.

SL's). Thus, there is a possibility of  $\zeta$  vanishing at a frequency in the vicinity of the  $m$ th-order Bragg frequency  $\omega_m = m\omega_1$ . Putting  $\zeta = \zeta_m + (\omega - \omega_m)\zeta'_m$  for  $\omega$  around  $\omega_m$ , the frequency  $\tilde{\omega}_m$  satisfying  $\zeta(\tilde{\omega}_m) = 0$  is evaluated as  $\tilde{\omega}_m = \omega_m - \zeta_m/\zeta'_m$ , where

$$\zeta_m \equiv \zeta(\omega_m) = (-1)^m \frac{\varepsilon}{2} \sin(2\omega_m d_A/v_A), \quad (10)$$

$$\zeta'_m \equiv \left. \frac{d\zeta}{d\omega} \right|_{\omega_m} = (-1)^{m+1} \left[ \frac{\pi}{\omega_1} + \varepsilon \left\{ \frac{d_A}{v_A} \sin^2(\omega_m d_A/v_A) + \frac{d_B}{v_B} \cos^2(\omega_m d_A/v_A) \right\} \right]. \quad (11)$$

Therefore, up to the order of  $\varepsilon$ , we have

$$\tilde{\omega}_m = \omega_m + \frac{\varepsilon}{2\pi} \omega_1 \sin(2\omega_m d_A/v_A). \quad (12)$$

On the other hand, the function  $F(\omega)$  defined by Eq. (3) depends weakly on  $\omega$  inside the frequency gap. Thus, we put  $F(\omega) \cong F(\omega_m) \equiv F_m$  in the  $m$ th frequency gap. Consequently, in the vicinity of  $\tilde{\omega}_m$ , Eq. (2) is approximated as

$$\Theta \cong 2 \tan^{-1} \left( \frac{F_m}{(\omega - \tilde{\omega}_m) \zeta'_m} \right), \quad (13)$$

and the phase time is expressed as a form characteristic of the resonance:

$$\tau = \frac{d\Theta}{d\omega} = \frac{2\Gamma_m}{(\omega - \tilde{\omega}_m)^2 + \Gamma_m^2}, \quad (14)$$

where the width  $\Gamma_m$  ( $\equiv -F_m/\zeta'_m$ ) of the resonance peak is expressed as

$$\Gamma_m = \frac{\omega_1}{\pi} \varepsilon \sin^2(\omega_m d_A/v_A) \left\{ 1 + \frac{\coth[N\varepsilon \sin(\omega_m d_A/v_A)]}{\sin(\omega_m d_A/v_A)} \right\}, \quad (15)$$

and we have chosen  $Z_A = Z_X$  for simplicity.

The frequency dependence of the phase time  $\tau$  calculated from the resonance formula Eq. (14) is compared in the inset of Fig. 2(a) with the numerical result. Here we note that  $\tilde{\omega}_2$  satisfying  $\zeta(\tilde{\omega}_2) = 0$  is found inside the second, narrow gap close to the lower edge. For the parameters we have chosen ( $d_A = d_B$  and  $v_A \cong v_B$ ),  $\sin(\omega d_A/v_A) \cong 0$  for a frequency  $\omega$  within this gap. This leads to the fact that the width of the frequency gap  $|\Delta_m| = (\omega_1/\pi) |\sin(\omega_m d_A/v_A)|$  becomes small for  $m=2$ . At the same time  $\coth[N\varepsilon \sin(\omega_2 d_A/v_A)]$  becomes very large though  $N$  is large ( $N=8$ ), leading to the width of the resonance much larger than the gap width, i.e.,

$$|\Gamma_m/\Delta_m| = |\sin(\omega_m d_A/v_A) + \coth[N\varepsilon \sin(\omega_m d_A/v_A)]| \gg 1, \quad (16)$$

for  $m=2$ . Thus, the enhancement of  $\tau$  is not found in the second frequency gap. It is readily seen that  $\varepsilon < 0$  (i.e.,  $Z_A > Z_B$ ) is necessary for  $|\Gamma_m/\Delta_m| \leq 1$ . In addition,  $|\coth[N\varepsilon \sin(\omega_m d_A/v_A)]| \leq 2$ , or  $|N\varepsilon \sin(\omega_m d_A/v_A)| \geq 1/2$  should be satisfied. This explains the fact that no peak of the phase time is seen inside the gaps when the stacking order of the bilayer is interchanged, i.e., the surface layer  $B$  is replaced with the  $A$  layer [see Fig. 2(b)].

Now we show that the displacement field of the phonons with the resonance frequency  $\tilde{\omega}_m$  is localized near the surface. In the continuum model, the lattice displacement and stress should be continuous at each interface of the adjacent layers. This boundary condition can be expressed as

$$\begin{pmatrix} U_{n+1} \\ S_{n+1} \end{pmatrix} = \begin{pmatrix} \mu & -\sigma/(\omega Z_A) \\ -\omega Z_A \zeta & \lambda \end{pmatrix} \begin{pmatrix} U_n \\ S_n \end{pmatrix}, \quad (17)$$

where,  $U_n$  and  $S_n$  are the displacement and stress at the  $n$ th interface, respectively. The elements  $\zeta, \mu$ , and  $\lambda$  of the ‘‘transfer matrix’’ are given by Eqs. (4) to (6), and  $\sigma$  is defined by

$$\sigma = \sin k_A d_A \cos k_B d_B + \frac{Z_A}{Z_B} \cos k_A d_A \sin k_B d_B. \quad (18)$$

For a SL with a free surface, an additional boundary condition is imposed, i.e., the stress at the surface should vanish, or  $S_0 = 0$  ( $n=0$  denotes the surface), see Fig. 1. With the use of this condition together with the equation  $\zeta = 0$  satisfied by the phonons of frequency  $\tilde{\omega}_m$ , Eq. (17) leads to

$$U_n = [\mu(\tilde{\omega}_m)]^n U_0, \quad S_n = 0, \quad (n=0, 1, 2, \dots), \quad (19)$$

where

$$\mu(\tilde{\omega}_m) = \frac{\cos(\tilde{\omega}_m d_B/v_B)}{\cos(\tilde{\omega}_m d_A/v_A)}. \quad (20)$$

Thus, if  $|\mu| < 1$ , the frequency  $\tilde{\omega}_m$  corresponds to a vibration localized near the surface, whose displacement decays exponentially with the distance away from the surface. Since the function  $\mu$  depends only weakly on  $\omega$  inside a frequency gap, we can approximate

$$\mu(\tilde{\omega}_m) \cong \mu(\omega_m) = (-1)^m [1 + \varepsilon \sin^2(\omega_m d_A/v_A)]. \quad (21)$$

This equation implies that the surface mode exists if  $\varepsilon < 0$ , i.e.,  $Z_A > Z_B$  but not for  $Z_A < Z_B$ . Hence, the existence of the surface vibrations is a necessary condition for the resonance enhancement of time delay of reflected phonons. We also note that in the second gap  $|\mu(\omega_m)|$  is substantially equal to unity because of  $\sin^2(\omega_m d_A/v_A) \cong 0$ , whereas  $|\mu(\omega_m)| \cong 1 + \varepsilon$  in the first and third gaps. Accordingly, at  $\omega = \tilde{\omega}_2$  the amplitude decays very slowly and the corresponding localized vibration behaves rather like a bulk mode in a frequency band. This also explains why the enhancement of  $\tau$  cannot be seen in the second frequency gap.

For a semi-infinite SL system, Camley *et al.*<sup>8</sup> derived the equation  $\zeta(\omega) = 0$  giving the frequencies of surface modes and the expression of the decay parameter  $\mu$ . Their result is essentially the same as our results. Equations (19) and (20) are valid for both the finite and semi-infinite SL systems.

In order to see explicitly the time delay of the reflected phonons, we examine the time development of a phonon wave packet whose average wave number in the initial state is  $k_m = \tilde{\omega}_m/v$ , where  $v \equiv v_X$ . As an initial packet, we assume a Gaussian of the form of

$$\psi_i(x, 0) = \exp \left[ -\frac{(x-x_0)^2}{4(\Delta x)^2} + ik_m x \right], \quad (22)$$

where  $x_0$  is the coordinate at the center of the packet. The Fourier component  $\phi(k)$  of this packet is given by

$$\phi(k) = \frac{2\sqrt{\pi}}{\Delta k} \exp \left[ -\frac{(k-k_m)^2}{(\Delta k)^2} + i(k_m - k)x \right], \quad (23)$$

where the magnitude of  $\Delta k \equiv 1/(\Delta x)$  is chosen so that  $\phi(k)$  is finite only inside the frequency window corresponding to the  $m$ th frequency gap. If the reflected phonon packet is well separated from the superlattice, it can be written as

$$\begin{aligned} \psi_r(x, t) &\cong \int \frac{dk}{2\pi} \phi(k) r(k) e^{-i(kx + \omega t)} \\ &= \int \frac{dk}{2\pi} \phi(k) e^{-i(kx + \omega t + \Theta)}. \end{aligned} \quad (24)$$

The intensity  $I_r = |\psi_r|^2$  calculated numerically from Eq. (24) supplemented by Eqs. (2) to (8) is plotted in Fig. 3 together with the initial wave packet. In this calculation, we choose  $x_0 = 0$ ,  $m = 1$  (the lowest gap), and  $\Delta\omega = v\Delta k = 10$  GHz. The reflected packets are illustrated as functions of  $x + vt$ . From Fig. 3, we see the spatial delay of the packet which is

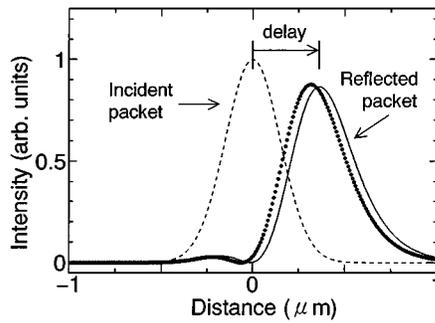


FIG. 3. Asymptotic forms of the reflected intensities of the phonon wave packet calculated from the exact expression Eq. (24) (solid line) and approximate expression Eq. (25) (dots) which are compared to the intensity of the initial Gaussian packet (dashed line). The reflected packets are illustrated as functions of  $x + vt$ .

equivalent to the corresponding time delay. An interesting feature is that the reflected packet has double peaks but the former peak is much smaller than the latter one. The similar double-peak structure of the reflected packet has also been obtained for the transmission of phonons through a double barrier structure. However, in this case the former peak is much larger than the latter peak. Now, the distance between the peaks of the initial packet and the latter large peak of the reflected packet gives the time delay.

For the approximated formula of  $\Theta$  given by Eq. (10), the integral in Eq. (24) can be analytically performed, leading to

$$\psi_r(x, t) = e^{-ik_m x_r} \left[ \exp\left(\frac{(x_r \Delta \omega)^2}{v^2}\right) - \frac{2\sqrt{\pi}\Gamma_m}{\Delta \omega} \times \exp\left(\frac{\Gamma_m^2}{(\Delta \omega)^2} - \frac{\Gamma_m x_r}{v}\right) \operatorname{erfc}\left(\frac{\Gamma_m}{\Delta \omega} - \frac{x_r \Delta \omega}{2v}\right) \right], \quad (25)$$

where  $x_r = x + vt$ . The intensity  $I_r = |\psi_r|^2$  calculated from Eq. (25) is also plotted in Fig. 3 by dots. The small deviation from the numerical result (solid line) is due to the fact that we consider only the effect of the resonance and it comes from the difference between the phase times calculated from the exact and approximate expressions illustrated in Fig. 2(a).

In the present work, we have shown that the phase associated with the reflected amplitude describes the temporal delay for the phonon packet scattered off a finite-size SL system. This delay is caused as a result of the interaction of incident phonons with a surface vibrational mode. In the numerical example shown in Figs. 2(a) and 3, the time delay at a resonant frequency becomes as large as 0.1 ns for the system of about 70 nm thickness. This means that the magnitude of the time delay of reflected phonons associated with the resonant interaction with a surface vibrational mode is in the range detectable by a phonon experiment using picosecond laser technique.<sup>22</sup> The concept of the phase time is an old one but it proves to be also useful for studying the existence of surface vibrational modes. We have shown that this phase time has large resonant peaks at eigenfrequencies of the surface modes in SL's, though the reflection rate is exactly unity for the whole range of frequency. Our results suggest the observability of the surface vibrational modes by a time-resolved phonon reflection experiment.

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