Time-resolved study of vibrations of *a*-Ge:H/*a*-Si:H multilayers

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We describe experiments in which the vibrations of *a*-Ge:H/*a*-Si:H multilayer structures on silica substrates are studied in real time. The vibrations are generated when a picosecond light pulse is absorbed in the structure, thereby setting up an elastic stress. The resulting motion is studied through a measurement of the change in the optical reflectivity as a function of time. We analyze the results in terms of the spectrum of normal modes of the multilayers and show that there exist surface modes at the free surface of the multilayer.

I. INTRODUCTION

Multilayer structures (crystalline or amorphous) have interesting vibrational properties that have been studied by Raman[1,2] and phonon transmission spectroscopy.[3,4] The Raman studies[1,2] confirm the theoretically expected folding of the acoustic branch of the phonon spectrum. Phonon transmission spectroscopy[3,4] gives clear evidence of band gaps, i.e., frequency intervals in which phonon propagation cannot occur. In this paper we use a new method which enables us to study in the time domain the vibrations of a multilayer structure (or any other microstructure). We will show that this new method gives extra, and interesting, information about the modes of the multilayer, particularly the surface modes.

II. EXPERIMENTAL METHOD

The basic idea of our method is shown schematically in Fig. 1. The vibrations of the multilayer structure are excited when a short light pulse (the "pump" pulse) is absorbed. As a light source we used a passively mode-locked colliding-pulse ring dye laser[5] operating at 6180 Å (2 eV). The pulse width is 0.1 psec, the energy per pulse 0.2 nJ, and the repetition rate 108 MHz. When the light pulse is absorbed in the multilayer a stress distribution is set up, and the structure is set into vibration. According to the simplest picture of this process we can imagine that the absorption of the light raises the temperature, and so the stress is just due to thermal expansion. In fact, as we have shown in earlier work,[6] there may also be an electronic contribution to the stress which is of the same order of magnitude as the thermal stress. Because of the different optical properties of Ge and Si, the spatial dependence of the initial stress distribution is rather complicated. We will discuss the theory of this in a later section.

When the multilayer structure begins to vibrate, there is a time-varying elastic strain in each layer. The strain changes the optical constants (n and α), and this, in turn, leads to a variation of the optical reflectivity of the structure with time. In our experiment we use a measurement of this change in reflectivity, $\Delta R(t)$, to study the vibrational modes. We measure $\Delta R(t)$ by means of a time-delayed optical probe pulse (pump-and-probe method). This probe has the same characteristics as the pump pulse and the reflected probe pulse is detected by a photodiode. To improve the signal-to-noise ratio the pump-pulse chain was modulated at 4 MHz, and the photodiode output fed into a phase-sensitive amplifier locked to this modulation frequency. The observed changes in reflectivity had magnitudes in the range 10^{-6}–10^{-4}.

The multilayers were prepared by plasma-assisted chemical-vapor deposition.[7] The ratio of the silicon layer thickness to the germanium layer thickness was kept constant at 1.13 to 1. The repeat distance d_r covered the range 19–1134 Å. The first and last layers were always silicon, but they did not necessarily have the same thickness as the silicon layers inside the multilayer. The total thickness of the multilayers varied between 0.5 and 1 μm. The number of bilayers varied from 5.5 for $d_r = 1134$ Å

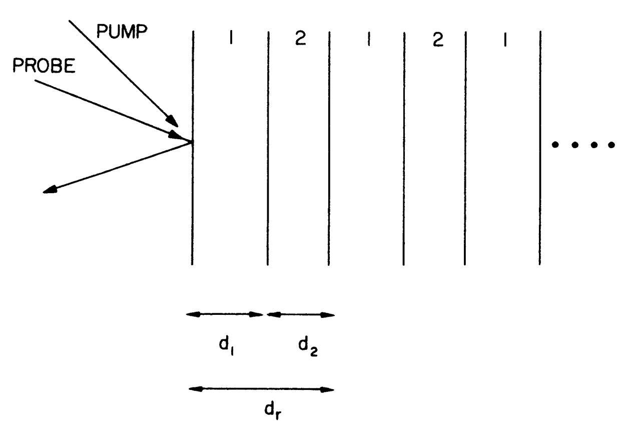

FIG. 1. Schematic diagram of the experimental configuration. Layers 1 and 2 refer to Si and Ge, respectively.

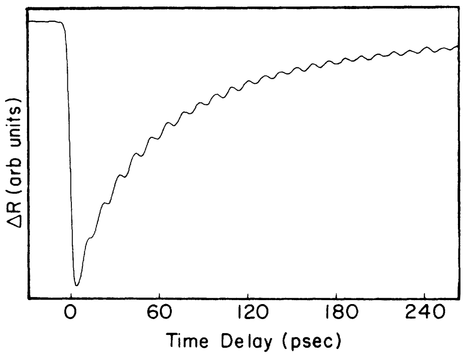

FIG. 2. Photoinduced reflectivity changes observed in a multilayer with a germanium layer thickness d_G of 190 Å and a silicon layer thickness d_S of 215 Å.

to more than 100 for the smallest repeat distance.

In Fig. 2 we show as typical results the response $\Delta R(t)$ for a multilayer with $d_r = 405$ Å. It consists of two superimposed terms. The first term is a rapid decrease of ΔR immediately after $t=0$ followed by a decay. The second term consists of a weakly damped oscillation of frequency 91.3 GHz, together with some irregular oscillatory components which persist only for a short time. (The existence of these components is not evident to the eye from Fig. 2, but we mention them at this point for the sake of completeness.) The first term is qualitatively similar to results we have obtained in studies[6] of a single film of a-Ge:H. It is the result of the change in optical properties due to photoexcited carriers. The second term (oscillatory component) is clearly a result of the vibration of the multilayer structure.

III. THEORY OF MULTILAYER EIGENMODES

In this section we study the normal modes of a multilayer structure. We consider a multilayer structure made up of alternating layers of thicknesses d_1 and d_2, densities ρ_1 and ρ_2, and sound velocities v_1 and v_2. The repeat distance d_r equals $d_1 + d_2$. In our experiments the diameter of the region which is illuminated is roughly 10 μm, and is therefore much larger than the thickness of the multilayer structure. Consequently, we only to need consider motion of the multilayer in the direction normal to the layers, i.e., in the z direction.

A. Modes of an infinite multilayer

The equation of motion is

$$\rho \frac{\partial^2 u(z,t)}{\partial t^2} = \frac{\partial \sigma}{\partial z}(z,t) , \quad (1)$$

where u is the displacement in the z direction, and σ is the zz component of the elastic stress tensor which is related to u by

$$\sigma(z,t) = 3B \frac{1-\nu}{1+\nu} \eta(z,t) , \quad (2)$$

where B is the bulk modulus, ν is Poisson's ratio, and η is the zz component of the elastic strain tensor ($=\partial u / \partial z$). Combining (1) and (2) gives the usual result for the sound velocity v,

$$v^2 = \frac{3B}{\rho} \frac{1-\nu}{1+\nu} . \quad (3)$$

We look for a solution in the form

$$u(z,t) = w(z) e^{-i\omega t} . \quad (4)$$

Let the mth layer of the multilayer begin at $z = z_m$. We can write the general solution of the wave equation in this layer in the form

$$w(z) = A_m \sin[k_m(z - z_m)] + B_m \cos[k_m(z - z_m)] , \quad (5)$$

where A_m and B_m are amplitudes, and k_m is equal to ω / v_1 or ω / v_2 depending on the layer. At the interface between two layers the displacement and the stress must be continuous. These conditions relate A_{m+1} and B_{m+1} to A_m and B_m. To determine the dispersion relation of the layered medium, we use a transfer matrix T defined by

$$\begin{bmatrix} A_{m+2} \\ B_{m+2} \end{bmatrix} = T \begin{bmatrix} A_m \\ B_m \end{bmatrix} . \quad (6)$$

The elements of T are

$$T_{11} = \cos(k_1 d_1)\cos(k_2 d_2) - p^{-1}\sin(k_1 d_1)\sin(k_2 d_2) , \quad (7)$$

$$T_{12} = -\sin(k_1 d_1)\cos(k_2 d_2) - p^{-1}\cos(k_1 d_1)\sin(k_2 d_2) , \quad (8)$$

$$T_{21} = \sin(k_1 d_1)\cos(k_2 d_2) + p\cos(k_1 d_1)\sin(k_2 d_2) , \quad (9)$$

$$T_{22} = \cos(k_1 d_1)\cos(k_2 d_2) - p\sin(k_1 d_1)\sin(k_2 d_2) , \quad (10)$$

where $p = \rho_1 v_1 / \rho_2 v_2$. These elements are based on the assumption that layer m is a type-1 layer. From (7)–(10) we find that $\det T = 1$, and hence the eigenvalues of T must be expressible in the form $(e^{i\theta}, e^{-i\theta})$. Then it is natural to define the wave number q by the relation

$$\theta = q d_r . \quad (11)$$

The trace of T is

$$T_{11} + T_{22} = e^{i\theta} + e^{-i\theta} = 2\cos(q d_r) . \quad (12)$$

This gives the dispersion relation

$$\cos(q d_r) = \cos(\omega d_1 / v_1)\cos(\omega d_2 / v_2) - \frac{(1+p^2)}{2p}\sin(\omega d_1 / v_1)\sin(\omega d_2 / v_2) , \quad (13)$$

in agreement with the result first derived by Rytov.[8] The folding of the acoustic branch of the spectrum in GaAs/$Ga_{1-x}Al_xAs$ superlattices has been shown to be well described by this formula.[9] In Fig. 3 we show the calculated dispersion curve for a Ge/Si multilayer with Ge and Si layer thicknesses of 200 and 226 Å, respective-

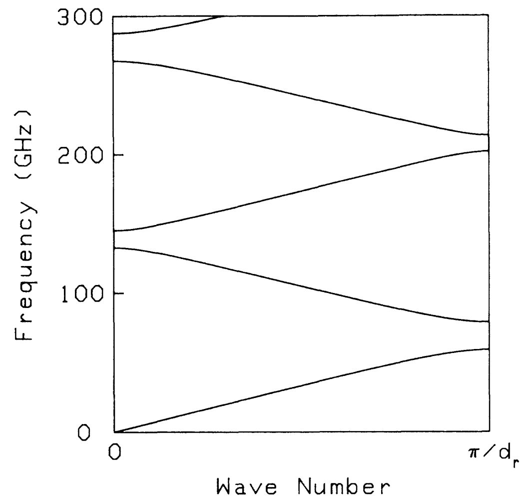

FIG. 3. Calculated dispersion relation for longitudinal-acoustic phonons in a Ge/Si multilayer with $d_G = 200$ Å and $d_S = 226$ Å. d_r is the repeat distance of the multilayer. The densities and sound velocities are given in the text.

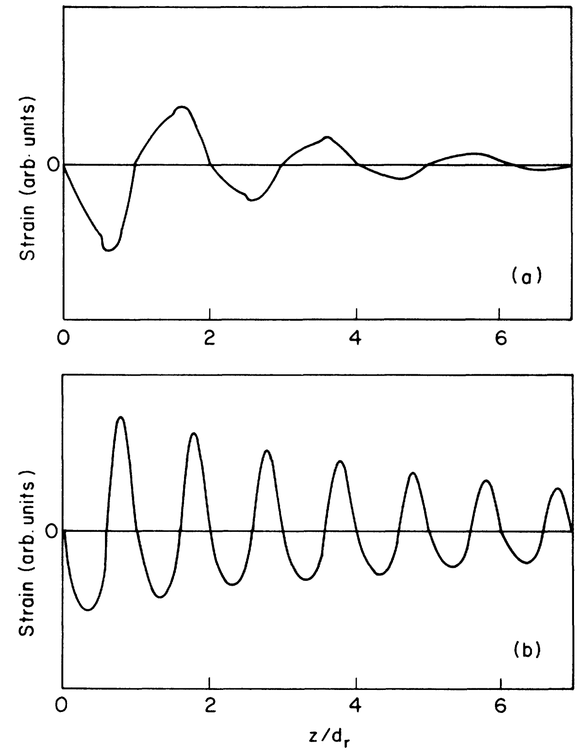

FIG. 4. Spatial dependence of the strain for the surface modes in (a) the lowest zone-boundary and (b) the lowest zone-center gap for a Ge/Si multilayer beginning with a Si layer. d_r is the repeat distance of the multilayer. Parameters are as given in the text.

ly ($d_S/d_G = 1.13$). The density and sound velocity in these materials are not accurately known and depend upon the conditions of preparation.[6,10–12] For the moment, we take the values $\rho_G = 4.85$ g cm^{-3}, $\rho_S = 1.76$ g cm^{-3}, $v_G = 4.6\times10^5$ cm sec^{-1}, and $v_S = 7.9\times10^5$ cm sec^{-1}. In Table I we list the phonon frequencies of the band edges.

B. Local modes at a free surface

Consider now a semi-infinite multilayer with a free surface at $z=0$. The first layer is composed of the material labeled 1. In this situation the spectrum may contain surface modes, in addition to the bulk modes of the infinite system. The theory of such modes has been discussed extensively, both for discrete and continuum models.[13] To derive the conditions for a surface mode let us suppose that the eigenvectors of T are $\phi^{(1)}$ and $\phi^{(2)}$. The pair of amplitudes A_1, B_1 in the first layer can be written as a linear combination of $\phi^{(1)}$ and $\phi^{(2)}$, viz.,

$$\begin{bmatrix} A_1 \\ B_1 \end{bmatrix} = C_1\phi^{(1)} + C_2\phi^{(2)} . \tag{14}$$

Then in the $(2n+1)$th layer the amplitudes are

$$\begin{bmatrix} A_{2n+1} \\ B_{2n+1} \end{bmatrix} = C_1\lambda_1^n\phi^{(1)} + C_2\lambda_2^n\phi^{(2)} , \tag{15}$$

when λ_1 and λ_2 are the eigenvalues of T, i.e., $e^{i\theta}$ and $e^{-i\theta}$. It is clear that for these amplitudes to become very small for large n we must have θ complex. If we choose $\text{Im}(\theta) > 0$, then $|\lambda_1| < 1$ and $|\lambda_2| > 1$. From (15) we

TABLE I. Frequencies of the lower (ν_-) and upper (ν_+) band edges and surface modes (ν_S) for the four lowest gaps for a Ge/Si multilayer with $d_G = 200$ Å and $d_S = 226$ Å. The eigenvalue λ_1 and the spatial decay length l in units of d_r are also listed. Parameters used in the calculation are given in the text.

Gap	ν_- (GHz)	ν_+ (GHz)	ν_S (GHz)	λ_1	l/d_r
1	59.4	79.0	72.6	−0.66	2.4
2	132.8	145.2	133.8	0.85	6.2
3	202.1	213.7	212.8	−0.87	7.2
4	267.6	287.5	274.7	0.65	2.3

then need $C_2=0$, which means that the amplitudes (A_1,B_1) in the first layer must be an eigenvector of T corresponding to the eigenvalue λ_1. An additional requirement is the condition that the stress vanishes at the free surface. From (5) we see that this gives $A_1=0$. Hence, the conditions for a surface mode are that the transfer matrix has an eigenvector (0,1), and the eigenvalue associated with this eigenvector must have a magnitude less than 1. The eigenvector requirement is equivalent to the condition $T_{12}=0$, which gives[14]

$$p\tan(\omega d_1/v_1)+\tan(\omega d_2/v_2)=0 . \quad (16)$$

The eigenvalue condition is

$$|\lambda_1|=\left|\frac{\cos(\omega d_1/v_1)}{\cos(\omega d_2/v_2)}\right|<1 . \quad (17)$$

If $p<1$ it is straightforward to show that for all frequencies which are solution of Eq. (16) $|\lambda_1|$ is <1, and so Eq. (17) is unnecessary in this case. On the other hand, if $p>1$, $|\lambda_1|$ is always >1, and so there are never any surface modes. Thus, surface modes only exist if the first layer of the multilayer has a smaller acoustic impedance $(\rho_1 v_1)$ than the second. The eigenvalue is related to the exponential decay length l of the mode by

$$l=-d_r/\ln(|\lambda_1|) . \quad (18)$$

We have calculated the frequencies of the surface modes from Eq. (16) for a Ge/Si multilayer using the parameters given above. For a semi-infinite multilayer starting with a Si layer, we find that each gap contains one localized surface mode. The frequencies of these modes, their eigenvalues, and corresponding decay lengths are listed in Table I. The closer the surface-mode frequency is to a band edge, the larger the decay length l, as expected on physical grounds. The location of the frequencies within the band gap varies irregularly from band gap to band gap.

In Fig. 4 we show the spatial dependence of the strain associated with the surface modes in the lowest two band gaps. For the surface mode in the lowest zone-boundary gap the strain changes sign after each bilayer. In the first gap at the zone center the surface mode has the same sign after each bilayer.

The spectrum of a semi-infinite multilayer in which the first layer is germanium does not have any surface modes, because $\rho_G v_G$ is greater than $\rho_S v_S$, and hence $p>1$. Given the result, it is clear that if we start from a multilayer in which the first layer is germanium and then add an increasingly thick layer of silicon at some point surface modes must appear. It is straightforward to show, again using the transfer-matrix technique, that the frequencies are a solution of[15]

$$\tan\left[\frac{\omega d_2}{v_2}\right]+p\tan\left[\frac{\omega d_1}{v_1}\right]\left[\cos^2\left[\frac{\omega d}{v_2}\right]+\frac{1}{p^2}\sin^2\left[\frac{\omega d}{v_2}\right]-\left[\frac{1-p^2}{p^2}\right]\tan\left[\frac{\omega d^2}{v_2}\right]\sin\left[\frac{\omega d}{v_2}\right]\cos\left[\frac{\omega d}{v_2}\right]\right]=0 . \quad (19)$$

The eigenvalue is

$$\lambda_1=\cos\left[\frac{\omega d_1}{v_1}\right]\cos\left[\frac{\omega d_2}{v_2}\right]-p\sin\left[\frac{\omega d_1}{v_1}\right]\sin\left[\frac{\omega d_2}{v_2}\right]$$
$$-\frac{1}{p}\tan\left[\frac{\omega d}{v_2}\right]\left[\sin\left[\frac{\omega d_1}{v_1}\right]\cos\left[\frac{\omega d_2}{v_2}\right]+p\cos\left[\frac{\omega d_1}{v_1}\right]\sin\left[\frac{\omega d_2}{v_2}\right]\right] . \quad (20)$$

The thickness of the surface layer of silicon is d. In Fig. 5 we show the frequency and the damping length of the zone-boundary gap mode as a function of the silicon surface layer thickness. The surface mode exists for d in the range $d_+^{(1)}=0.5d_S$ to $d_-^{(1)}=2.0d_S$ (d_S is the thickness of the silicon layers in the interior of the multilayer), and reappears and disappears repeatedly as d is increased. The physics of this process is very simple. When $d=d_+^{(1)}$ the mode has a frequency equal to the frequency ω_+ at the upper limit of the band gap. Thus the wavelength in the silicon is

$$\lambda_+=\frac{2\pi v_2}{\omega_+} . \quad (21)$$

Hence, when

$$d=d_+^{(n)}\equiv d_+^{(1)}+\frac{n\lambda_+}{2} , \quad (22)$$

where n is any integer, one has a surface mode which is similar to the mode for $d=d_+^{(1)}$, but which has $n/2$ extra wavelengths in the silicon surface layer. This mode disappears when d is increased to

$$d_-^{(n)}=d_-^{(1)}+\frac{n\lambda_-}{2} , \quad (23)$$

where

$$\lambda_-=\frac{2\pi v_2}{\omega_-} \quad (24)$$

and ω_- is the frequency at the bottom of the gap. The fact that λ_+ is less than λ_- means that when d is sufficiently large there may be more than one mode in the gap.

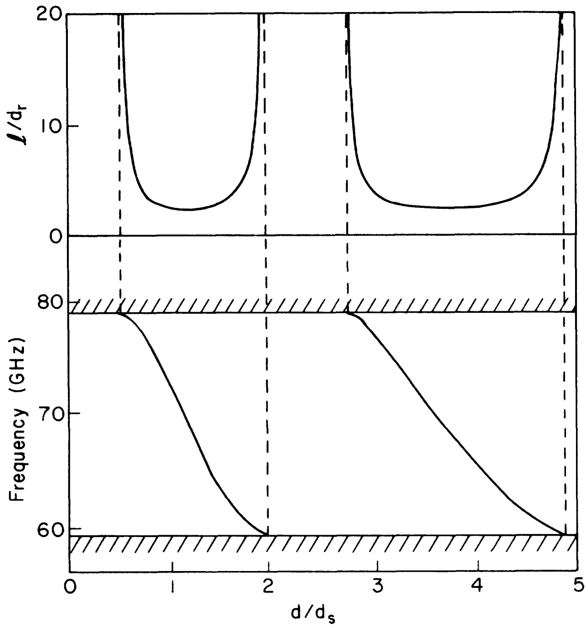

FIG. 5. Frequency and damping length l of the lowest zone-boundary gap mode as a function of the silicon surface layer thickness d. The thickness of the silicon layers in the interior of the multilayer is d_S, and d_r is the repeat distance of the multilayer. Parameters are as given in the text.

IV. COMPUTER SIMULATIONS

In this section we describe computer simulations which predict the form of $\Delta R(t)$ to be expected in the experiment. This involves a calculation of the spatial dependence of the stress produced when the pump light pulse is absorbed, computation of how the resulting strain in the multilayer develops as a function of time, and the change in the optical reflectivity caused by this strain.

A. Stress generation

We consider the experimental configuration shown in Fig. 1. The effective absorption length is assumed to be much smaller than the total thickness of the multilayer. The energy $W(z)$ deposited per unit volume at a distance z from the surface of the multilayer is

$$W(z)=\frac{\alpha(z)n(z)Q}{A}\left|\frac{E(z)}{E_0}\right|^2 , \quad (25)$$

where α and n are the absorption coefficient and refractive index, Q is the pulse energy, A is the illuminated area, and $E(z)$ and E_0 are the electric fields at distance z in the multilayer and of the incident light wave, respectively. As before, we can consider all quantities to depend just on the space variable z. The field amplitude $E(z)$ is calculated in a straightforward way by using the transfer-matrix method applied to electromagnetic waves. One can show that

$$E(z)=E_0[M_{11}+M_{21}+r_0(M_{12}+M_{22})] . \quad (26)$$

The matrix $M(z)$ is a transfer matrix for traveling light waves (as distinct from standing waves used in Sec. III A) between a point just outside the surface of the multilayer and the point z, and r_0 is the amplitude reflection coefficient of the multilayer.

For the optical properties of amorphous germanium and silicon, we used the values $\alpha_G=3.3\times10^5$ cm^{-1}, $n_G=5$,[16] $\alpha_S=2.5\times10^4$ cm^{-1}, and $n_s=4.5$.[17] In Fig. 6 we show the deposited energy as a function of z for a multilayer with $d_G=200$ Å. Since $\alpha_G \gg \alpha_S$ most of the energy is deposited in the germanium layers. The deposited energy leads to a temperature rise in the multilayer which sets up a thermal stress (isotropic stress tensor) of magnitude

$$\sigma(z)=\frac{-3B\beta W(z)}{C} , \quad (27)$$

where β is the linear expansion coefficient and C is the specific heat per unit volume. As noted earlier, there may also be an electronic contribution to the stress. However, this is unlikely to change the spatial distribution of the stress in a significant way, i.e., the stress will be set up primarily in the germanium layers, and will fall off with distance at a rate determined by α_G.

B. Sensitivity function

The oscillating elastic strain in the multilayer causes a change $\Delta R(t)$ of the optical reflectivity. Since the strain is always small, we can define a "sensitivity function" $f(z)$ such that

$$\Delta R(t)=\int f(z)\eta(z,t)dz . \quad (28)$$

The problem is to calculate $f(z)$ for a multilayer structure. Let N_m be the transfer matrix (for traveling electromagnetic waves) of the mth bilayer. Then, the optical reflectivity of a semi-infinite multilayer is

$$R=\lim_{n\to\infty} |(N_n\cdots N_2N_1)_{21}/(N_n\cdots N_2N_1)_{22}|^2 . \quad (29)$$

Now suppose that the strain has a form

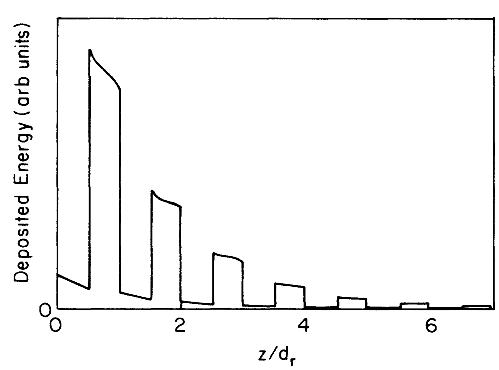

FIG. 6. Energy deposited by the pump pulse as a function of the distance z into a Ge/Si multilayer with $d_G=200$ Å and $d_S=226$ Å. Other parameters are given in the text.

$$\eta(z)=\delta(z-z') , \quad (30)$$

where z' lies in the mth bilayer. This causes a discontinuity in the optical properties at z', and an amplitude coefficient r' at this point given by

$$r'=\frac{2\pi i}{\lambda}\left[\frac{\partial n}{\partial \eta}+i\frac{\partial \kappa}{\partial \eta}\right] , \quad (31)$$

where λ is the light wavelength in vacuum, and κ is $\alpha\lambda/4\pi$. It is then straightforward to calculate (to first order in r') the change δN_m in the transfer matrix of the mth bilayer which contains the point z'. The change in R can then be found by differentiation of Eq. (29), and this change is $f(z')$ [see Eq. (28)].

For the purposes of this paper we only need the spatial dependence of $f(z)$, not the absolute magnitude. In other experiments performed previously,[6,18] we have found that in a-Si:H the strain-induced changes in the optical reflectivity are much smaller than in other materials (this is presumably because of the small value[19] of $\partial E_g/\partial P$ for a-Si:H). We therefore assume that $\partial n/\partial\eta$ and $\partial\kappa/\partial\eta$ are negligibly small in silicon. To determine the spatial dependence of $f(z)$, it is then sufficient to know the ratio of $\partial n/\partial\eta$ to $\partial\kappa/\partial\eta$ for germanium. For this ratio we use the model[20] described in Ref. 6. An example of the sensitivity function obtained in this way is shown in Fig. 7 for a multilayer with $d_G=200$ Å. When the repeat distance is very small compared to the absorption length in germanium, the range of the sensitivity function extends over many bilayers; for large repeat distances, on the other hand, the change in optical reflectivity is determined entirely by the strain in the first germanium layer. Depending on the layer thickness, the sensitivity function can be positive or negative in a particular layer, or it can change sign within the layer.

C. Calculation of ΔR

To calculate $\Delta R(t)$ for a particular multilayer, we first calculated the initial stress $\sigma(z)$. We assumed that this stress was set up instantaneously. The initial strain is zero. The equations of elasticity (1) and (2) were then integrated numerically to give the strain at later times, and $\Delta R(t)$ was found from Eq. (28). As an example, we show $\Delta R(t)$ for a multilayer with $d_G=200$ Å in Fig. 8(a). The response is dominated by an undamped oscillatory response with a frequency of 72.1 GHz. This is the frequency of the surface mode in the lowest zone-boundary gap (see Table I). There is also extra structure in $\Delta R(t)$, which becomes less important as time increases. This extra structure is due to the bulk modes of the multilayer. Since these have a continuous spectrum (apart from the band gaps), when they are excited they produce a response which may be oscillatory, but which will decay with time. As time progresses these bulk modes become increasingly out of phase with each other, and so interfere destructively. To illustrate this effect we show in Fig. 8(b) a simulation for a Ge/Si multilayer that begins with a germanium layer ($d_G=200$ Å and $d_S=226$ Å). According to the results of Sec. III B, surface modes do not exist for this configuration. The simulated $\Delta R(t)$ shows that the surface mode in the lowest zone-boundary gap has completely disappeared, but a damped oscillatory response with a frequency close to the lower band edge of the first zone-center gap is visible under these conditions. This response is due to the dephasing bulk modes.

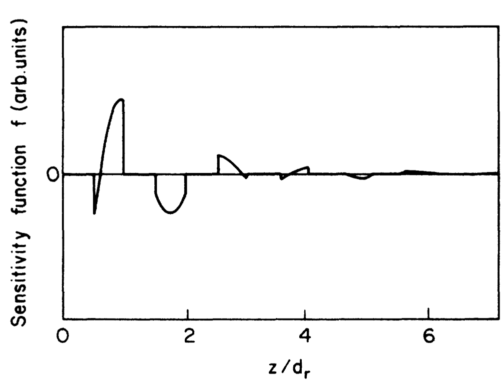

FIG. 7. Sensitivity function $f(z)$ for a Ge/Si multilayer with $d_G=200$ Å and $d_S=226$ Å. Other parameters are given in the text.

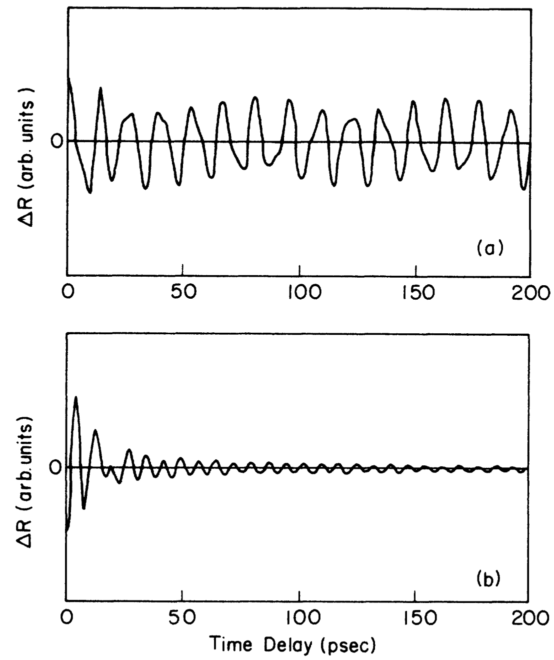

FIG. 8. Computer simulation of $\Delta R(t)$ for a Ge/Si multilayer with $d_G=200$ Å and $d_S=226$ Å beginning with (a) a silicon layer and (b) a germanium layer. Other parameters are given in the text.

V. RESULTS AND DISCUSSION

The characteristics of the samples studied are listed in Table II. The accuracy of the layer thickness is estimated

TABLE II. Characteristics of the samples studied. d_G is the germanium layer thickness. d/d_S is the ratio of the thickness of the first silicon layer to the silicon layer thickness in the interior of the multilayer. ν_{expt} is the experimentally observed frequency of the oscillations in $\Delta R(t)$ and ν_{theor} is the theoretical frequency discussed in the text.

d_G (Å)	Numbers of bilayers	d/d_S	ν_{expt} (GHz)	ν_{theor} (GHz)
89	50.5	1		163.0
134	34.5	0.97	124	109.0
190	19.5	0.6	91.3	82.8
534	5.5	1	32.3	27.2

to be ±5%. Results obtained for samples with d_G of 134, 190, and 534 Å are shown in Fig. 9. The smoothly varying background has been subtracted out (compare with Fig. 2). The change in reflectivity $\Delta R(t)$ is dominated by a persistent oscillatory response with frequency as listed in Table II. The observed frequencies indicate that the oscillations are due to the surface mode in the first zone-boundary gap. The theoretical frequencies are about 10–15% lower than the experimental results. This discrepancy is not surprising considering the uncertainty in the layer thickness, and the large variation of the sound velocity and density of a-Si and a-Ge depending on the preparation method.

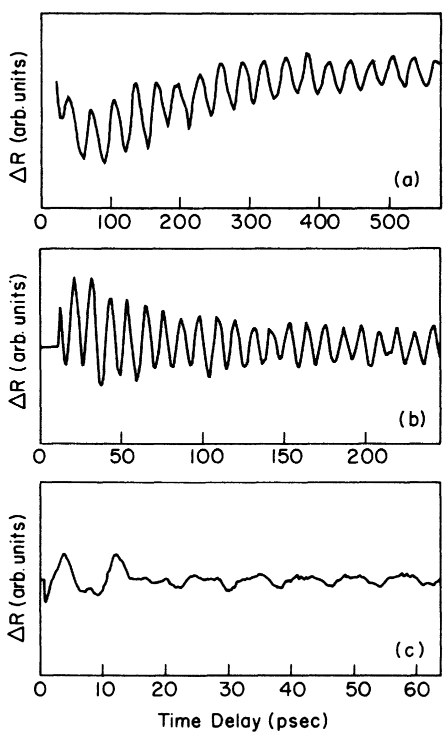

FIG. 9. Experimental results for ΔR as a function of time for Ge/Si multilayers. A smoothly varying background has been subtracted from the data. (a) $d_G = 534$ Å, (b) $d_G = 190$ Å, and (c) $d_G = 134$ Å.

For each of these three samples some damping of the oscillatory part of the response is observed. We believe, however, that the origin of this damping is different for different multilayers. The multilayer with $d_G = 534$ Å consists of only 5.5 bilayers, and consequently the surface mode still has an appreciable amplitude at the substrate interface. Thus, damping can occur due to radiation of sound into the substrate. We have performed a computer simulation for this multilayer on a silica substrate, and have found that the damping is roughly of the magnitude we observe.

For the two thinner multilayers the effect of leakage of the surface mode into the substrate is negligible. The damping in these multilayers is probably due to acoustic attenuation. For the 190-Å multilayer one can measure the decay time of the oscillations with reasonable accuracy, and the result is ~ 150 psec. This implies an acoustic attenuation of 0.06 dB psec^{-1} at the frequency of 91 GHz. This is very similar to the attenuation we have measured[21] in the same frequency range in a-SiO_2. For the 134-Å multilayer we cannot measure the attenuation because the oscillations are not so strong.

As can be seen from Fig. 9(c) the magnitude of $\Delta R(t)$ for $d_G = 134$ Å is very small. We have also measured ΔR for several multilayers with even smaller repeat distances. For $d_G = 89$ Å we can just manage to detect an oscillatory signal, but for $d_G = 60$ Å or less we can see no oscillations. We have examined several possible explanations for this. The first possibility is that because of the spatial form of the initial stress or the sensitivity function surface modes are not appreciably excited or detected. To test this idea we performed computer simulations of $\Delta R(t)$ for multilayers with $d_G = 134$ and 40 Å. For $d_G = 134$ Å we found that there should still be a strong signal due to the surface mode in the first zone-boundary gap. For $d_G = 40$ Å the dominant contribution is from the surface mode in the first zone-center gap, and the size of the oscillation is still of the same order of magnitude as for the thicker multilayers. Thus, this explanation is not sufficient to explain the absence of oscillations. It is also clear that one cannot explain the effect in terms of acoustic attenuation. From Fig. 9(c) we can see that the amplitude is very small, but nevertheless several oscillations of approximately the same amplitude occur.

We think a promising possibility is to consider at a more microscopic level the way the stress is generated by the incident pump pulse. In our calculations so far we have assumed that the stress is produced instantaneously and has a spatial distribution following the absorption profile of the light. If, in fact, the stress takes a certain time τ_σ to increase to its full value, the generation of high-frequency vibrations (such that $\omega\tau_\sigma \sim 1$) will be greatly reduced. For example, if we assume that σ has the time dependence

$$\sigma = \begin{cases} 0, & t<0 \\ \sigma_\infty \tanh(t/\tau_\sigma), & t>0 \end{cases} \tag{32}$$

one can show that the generation is reduced by a factor of 3 when $\omega\tau_\sigma \sim 4$. The zone-boundary surface mode for $d_G = 134$ Å has a frequency of 124 GHz. Thus, for this mechanism to reduce the amplitude by a factor of 3, we would need τ_σ to be ~ 5 psec. To understand at the microscopic level how the stress develops with time, one must consider how energy is transferred from the hot carriers to the thermal phonons (acoustic and optical). Initially, hot carriers lose their energy through emission of optical phonons (in ~ 1 psec), and these convert into acoustic phonons subsequently (time 3–10 psec). Thus, to explain a time constant for the stress of ~ 5 psec we would have to propose that the major contribution to the stress is not produced until the thermal acoustic phonons appear. This would require that in a-Ge and a-Si the acoustic thermal phonons have significantly larger Grüneisen constants than the optical phonons.[22]

The spatial variation of the initial stress may also be different from what we have assumed. The hot carriers excited by the pump pulse have a very large diffusion coefficient. For an excess energy of 0.5 eV the carrier velocity is 4200 Å psec^{-1} (if $m^*/m = 1$). If we assume a mean free path of 10 Å, the diffusion coefficient D is then 14 000 $\text{Å}^2\,\text{psec}^{-1}$. The carriers cool to close to the band edge in a time $\tau_c \sim 1$ psec, and therefore can diffuse a distance x given by

$$x = (D\tau_c)^{1/2} = 118 \text{ Å} . \tag{33}$$

Thus, in a multilayer with a small layer thickness some carriers can diffuse from the germanium where they are generated into the silicon, and can therefore deposit energy in the silicon. Hence, the spatial dependence of the deposited energy can be appreciably different from what we have assumed. The difference in the stress between adjacent germanium and silicon layers will be reduced by this diffusion, and this clearly will decrease the amplitude of the generated surface modes.

We have also measured the photoinduced reflectivity changes for the substrate-multilayer interface. The time dependence of $\Delta R(t)$ for this configuration is shown in Fig. 10 for a Ge/Si multilayer with $d_G = 190$ Å and $d_S = 215$ Å. The oscillatory response is now strongly damped with a time constant of about 10–20 psec. This is expected since energy can be radiated into the substrate. The frequency of the oscillations is very similar to the frequency of the free surface response of $\Delta R(t)$ as shown in Fig. 2. We also performed a computer simulation for this configuration with a result similar to the experimentally observed response.

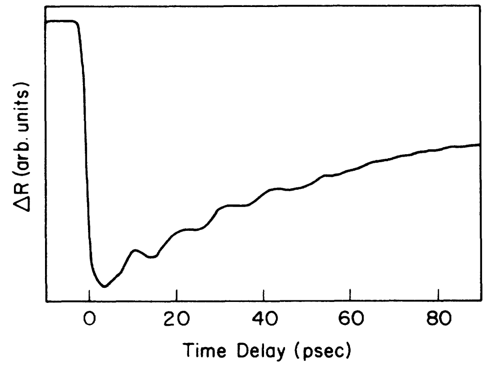

FIG. 10. Photoinduced reflectivity changes of the multilayer-substrate interface for a multilayer with $d_G = 190$ Å and $d_S = 215$ Å.

We have two additional pieces of evidence that the generation process is more complicated than we have assumed. The first of these comes from the details of $\Delta R(t)$ at short times, i.e., during the first few oscillations. We made a detailed experimental study of $\Delta R(t)$ during the first 30 psec for the multilayer with $d_G = 190$ Å. We have performed extensive computer simulations to try to reproduce the experimentally observed response. In these simulations we varied the sound velocities v_G and v_S, the thickness d_G (keeping d_S/d_G fixed), and the thickness d of the first silicon layer over a reasonable range. It was not possible to find parameters such that the simulated response was in agreement with the experimentally observed $\Delta R(t)$, suggesting that our assumption that the generation process is instantaneous may not be correct.

The second piece of evidence is the phase of the oscillations at long times, i.e., after 10 or more periods. We can write the contribution to $\Delta R(t)$ from the surface mode in the form

$$A \cos(2\pi\nu t - \phi) , \tag{34}$$

where the amplitude A is taken to be positive. It is straightforward to show that if the stress is produced instantly the phase ϕ must always be 0 or π. From the data shown in Fig. 9, one can determine the phase of the oscillatory signal for the multilayers with $d_G = 534$ and 190 Å. For the 534-Å sample the phase ϕ was found to be 0.97π, to within the experimental accuracy of $\pm 0.1\pi$. For the 190-Å sample ϕ was $\sim 1.5\pi$, which is consistent with a sudden turn on of the stress. To explain this we have to assume that there is a phase shift of $\sim 0.5\pi$ due to the finite time for the stress to be produced. Using the model in Eq. (32), we have calculated the phase shift as a function of $\omega\tau_\sigma$. If we assume $\tau_\sigma \sim 5$ psec (as required to explain the drop in the amplitude for the thin multilayers), the phase shift at a frequency of 90 GHz is 0.42π, and can thus explain the phase shift for the 190-Å sample. The phase shift at 32 GHz (surface-mode frequency in the 534-Å sample) is calculated to be 0.2π, which is not consistent with the experimental result. We are planning to carry out other experiments to test directly the hypothesis that there is time delay in the generation of the elastic stress.

VI. CONCLUSIONS

In this paper we have shown how to use the picosecond pump-and-probe technique to study the mechanical vibrations of microstructures. The technique has the advantage that the vibrations are time resolved, and so one is able to make measurements that are equivalent to observations of the motion of the structure in "real time." For the particular microstructures we have studied (multilayers of *a*-Ge:H/*a*-Si:H), the response of the structure to the pump light pulse is dominated by a surface mode polarized normal to the free surface of the structure. A study of this type of mode is of interest for two reasons. In the first place the frequency of the mode is a sensitive probe of the acoustic properties of the components of the microstructure, and of the geometry. We have shown, for example, that the frequency of the mode varies rapidly with the thickness d of the first layer of the multilayer, and that the mode disappears unless d lies in a certain range. The second point is that a measurement of the rate of damping of the surface-mode oscillations could be used to give accurate information about acoustic attenuation in the frequency range 10–100 GHz, i.e., above the upper limit of ordinary ultrasonic techniques. The advantage of the method is that since the mode is trapped at the surface there is a negligible damping due to acoustic radiation into the substrate (at least for multilayers with a sufficient number of bilayers). Of course, the measured attenuation is some combination of the attenuation in the two components of the multilayer. However, this is not a significant drawback if one is primarily interested in the temperature and frequency dependence of the attenuation in some class of materials (for example, amorphous semiconductors). We plan to make measurements of this type in the near future.

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[22]For a discussion of stress at a microscopic level, see Ref. 6.