Acoustoelastic anomaly in stressed heterostructures

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We investigated the influence of stress on the acoustic wave propagation in single crystalline heterostructures using a transfer matrix method. Both Rayleigh-type and Sezawa modes exhibit an acoustoelastic anomaly, where the stress-induced change of the phase velocity is maximum for finite film thicknesses, considerably smaller than the acoustic wavelength. For Ge/Si(001) compressed by 1 GPa the velocity shift of Sezawa modes reaches exceptionally high values of about 2%. These results demonstrate the importance of stress effects on the determination of elastic constants of thin film heterostructures.

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Ultrasonic waves are a unique tool for the nondestructive investigation of bulk materials, particularly for the precise determination of the elastic constants.[1] More recently ultrasonic waves have also been employed to measure the elastic properties of polycrystalline and epitaxial thin films.[2–5] Though still rare, such measurements are particularly important for heterostructures as the elastic constants play a key role for the understanding of the interdependence of stress and strain—with particular impact for electronic and optoelectronic devices. So far it is not clear whether the elastic constants of thin films deviate from the respective bulk values, which usually are used to calculate stress from strain and vice versa. Moreover, an increasing number of technologically interesting materials—such as cubic GaN or AlAs—are stable only in the thin film configuration and therefore require thin film measurements. For thin film investigations the higher surface sensitivity of surface acoustic waves (SAW's) is utilized, which have the vibration energy localized close to the surface, typically within a region of few wavelengths only. In the presence of a thin film SAW's become dispersive, i.e., the phase velocity depends on the frequency. Moreover, at certain material parameters and film thicknesses new surface guided modes, e.g., Love or Sezawa modes, arise.[6] Hence, a variety of different acoustic resonances are available to derive also the elastic constants of thin films with the necessary high accuracy.

It is well known from solid state acoustics that the phase velocity of acoustic waves is influenced by stress, a phenomenon called the acoustoelastic (AE) effect. Usually the relative change of the wave velocity is very small, e.g., $\simeq 10^{-5}$/MPa for aluminum.[7] Hence, stresses as high as 100 MPa typically applied in bulk experiments alter the phase velocity of the acoustic waves only by about 0.1%. In heteroepitaxial thin films, however, due to the misfit between film and substrate the residual stress can easily exceed the 1 GPa limit,[8] thus giving rise to phase velocity changes, which no longer are negligible.

For the stress dependence it is widely accepted that maximum change of phase velocity appears when the wave is completely localized within the stressed material. Consequently the wavelength of SAW's in layered structures has to be much smaller than the layer (i.e., film) thickness. In contrast to this presumption we recently found that the AE effect in stressed layered system assumes significant values for surface modes with penetration depths considerably larger than the film thickness.[9] For thin Ge films deposited on Si(001) the velocity change of Love modes increases rapidly with frequency and film thickness and reaches almost its maximum value when the wave is still penetrating deeply into the unstressed substrate. In 50 nm thick Ge films biaxially compressed by 1 GPa the velocity of the Love modes propagating parallel to the [110] direction changes by about 1% at 10 GHz operation frequency. Recent developments in high frequency SAW detection, in particular of surface Brillouin spectroscopy[10,11] and scanning acoustic probe microscopy[12] indeed promise an experimental accuracy of about 1%. Hence, the AE effect has to be carefully considered when recovering exactly the elastic properties of stressed systems by acoustic measurements.

In this study we calculated the influence of stress on the wave propagation in single crystalline heterostructures. Our calculations reveal an acoustoelastic anomaly, where the AE effect reaches its maximum value already at film thicknesses considerably smaller than the ultrasonic wavelength. For Rayleigh-type waves we find that the maximum velocity change occurs when the wave is not yet completely localized within the film. An analogous behavior is calculated for higher order Rayleigh modes, so called Sezawa modes, with the maximum velocity changes being even significantly higher.

The physical model has been explained in detail elsewhere.[9] The calculations are based on the prototype geometry shown in Fig. 1. A stressed layer is deposited onto a

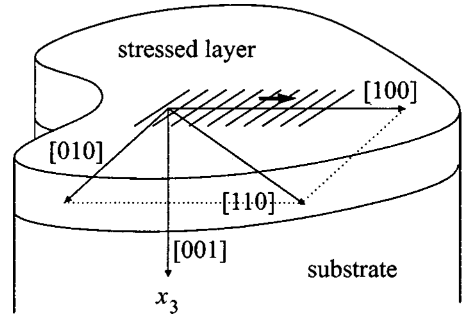

FIG. 1. Sketch of the SAW propagation geometry in a layered system. The $x_3 = 0$ plane is the interface between the layer and the substrate.

substrate. The $x_3=0$ plane of the Cartesian coordinate system is the interface between the layer and the substrate while the plane $x_3=-h$ is the free surface of the layer. In the following expressions ξ_i, u_{ij}, and σ_{ij} represent the particle displacement vector, the strain, and the stress, respectively. The notation without superscripts describes the acoustic variables and with subscript (0) the residual (static) ones. For the residual stresses we assume the biaxial model $\sigma_{11}^{(0)}=\sigma_{22}^{(0)}=\sigma^{(0)}$, $\sigma_{ij}^{(0)}=0$, for $i\neq j$. The residual strains are estimated by Hooke's law. Hence, the system contains only the axial strain components $u_{11}^{(0)}=u_{22}^{(0)}\neq 0$, $u_{33}^{(0)}\neq 0$. Then the wave propagation in the system can be described by equation of motion[7,9]

$$\frac{\partial \sigma_{ij}}{\partial x_j}+\sigma^{(0)}\frac{\partial^2 \xi_i}{\partial x_1^2}=\rho^{new}\frac{\partial^2 \xi_i}{\partial t^2}, \tag{1}$$

with the density

$$\rho^{new}\approx\rho(1-\Delta u^{(0)}),\quad \Delta u^{(0)}=u_{11}^{(0)}+u_{22}^{(0)}+u_{33}^{(0)} \tag{2}$$

modified by the static strain. The direction of wave propagation will be taken as the x_1 direction. Furthermore Hooke's law can be written as $\sigma_{ij}=C_{ijkl}(\partial\xi_k/\partial x_l)$ with C_{ijkl} being the modified stiffness modulus

$$C_{ijkl}=c_{ijkl}(1+u_{ii}^{(0)}+u_{jj}^{(0)}+u_{kk}^{(0)}+u_{ll}^{(0)}-u_{11}^{(0)}-u_{22}^{(0)}-u_{33}^{(0)}) + c_{ijklmn}u_{mn}^{(0)}, \tag{3}$$

with the summation performed over m,n where c_{ijkl} and c_{ijklmn} are the second- and third-order stiffness tensors, respectively.

Equations (1)–(3) represent the total system of equations for the wave propagation in a stressed body. For the surface wave propagation in a layered system additionally the boundary conditions at the interface between substrate and layer and at the free surface have to be fulfilled. Then the acoustic phase velocity was calculated by applying the modified transfer matrix approach described in Ref. 9.

The behavior of SAW in the (001) plane of cubic crystals have been investigated intensively before.[6] For SAW propagation along a cubic axis the sagittal plane, i.e., the plane determined by the wave vector and the surface normal, is a plane with mirror symmetry. Since only two particle displacements are involved, i.e., the vertical and the longitudinal components, and they are in phase quadrature, the surface displacement is elliptical and the wave is called a Rayleigh-type wave. For the propagation along the [110] direction there is also a wave solution having sagittal displacement only. However, this wave can be regarded as a singularity in that it begins to leak energy into the substrate when the propagation direction is slightly tilted from [110].

In the presence of a thin film with $\hat{v}_t<v_t$, where $\hat{v}_t$ and v_t is the transverse shear velocity for the bulk wave in the film and substrate, respectively, the film loads the substrate and the velocity of the free surface Rayleigh mode on the substrate material is decreased. Furthermore, when the film significantly loads the substrate,[6] higher-order Rayleigh modes, i.e., Sezawa modes, occur. Each of the Sezawa modes has a low-frequency cutoff at which the phase velocity is equal to the substrate shear velocity.

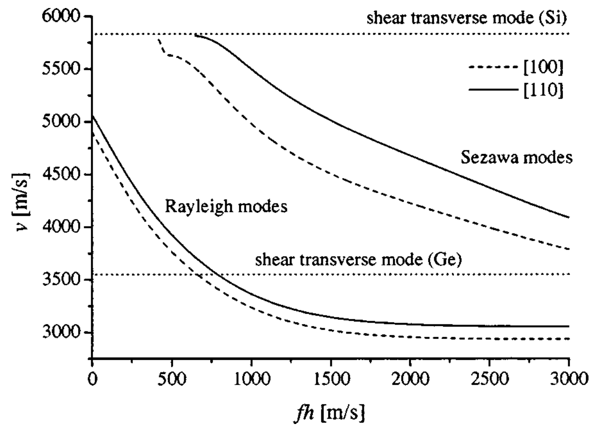

FIG. 2. Phase velocity dispersion v vs layer thickness h and frequency f of Rayleigh-type and Sezawa modes calculated for the unstressed Ge/Si(001) system. The Rayleigh mode starts at the Rayleigh velocity of Si and decrease with increasing fh until it becomes the Rayleigh velocity of Ge. The low frequency cutoff of the Sezawa mode is the shear transverse mode of Si and the high-frequency asymptote is the layer shear velocity.

In Fig. 2 the phase velocity dispersion of Rayleigh-type modes and Sezawa modes with propagation directions parallel to [100] and [110] in the unstressed layered Ge/Si(001) system are shown. For a very thin Ge film the Rayleigh wave is mainly determined by the elastic properties of Si. When the film thickness h or the operating frequency f increase the Ge layer slows down the velocity until eventually the Rayleigh-type wave is completely localized within the Ge film. The Sezawa mode requires a minimum layer thickness to propagate for a given frequency, e.g., at 10 GHz film thicknesses of about 40 nm and 60 nm are required for the [100] and [110] directions, respectively. Note that the surface and interface displacement describe retrogressive ellipses for Rayleigh waves and progressive ellipses for Sezawa waves. Furthermore, while the oscillation is mainly dominated by the vertical component for Rayleigh waves, for Sezawa waves the longitudinal amplitude may dominate in the Ge/Si(001) system. This is an important aspect for the understanding of the anomalous dependence of the AE effect discussed later.

For the calculation of the phase velocity and the penetration behavior we have used second-order elastic constants and density of Si ($c_{11}=165$ GPa, $c_{12}=64$ GPa, $c_{44}=79.2$ GPa, $\rho=2329$ kg/m^3), and Ge ($c_{11}=129$ GPa, $c_{12}=48$ GPa, $c_{44}=67.1$ GPa, $\rho=5323.4$ kg/m^3), as well as third-order elastic constants of Ge ($c_{111}=-720$ GPa, $c_{112}=-380$ GPa, $c_{123}=-30$ GPa, $c_{144}=-10$ GPa, $c_{155}=-305$ GPa, $c_{456}=-45$ GPa), respectively.[13] When the Ge film is biaxially stressed[14] the phase velocity dispersion changes slightly. Since this change is very small compared with the dispersion itself we introduce the phase velocity shift $\Delta v=v^{new}-v$, where v and v^{new} is the phase velocity for the system without and with residual stresses and strains, respectively.

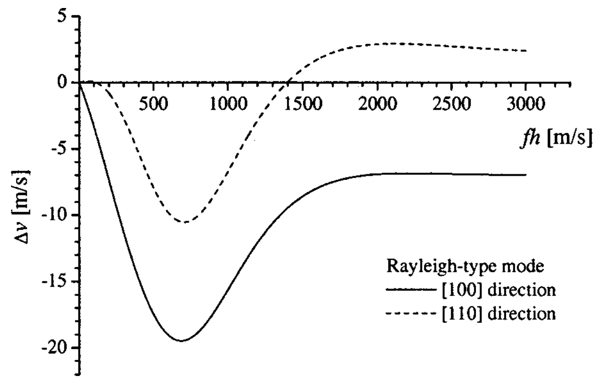

FIG. 3. Phase velocity change Δv vs frequency-thickness product fh for Rayleigh-type waves propagating parallel to the [100] and [110] direction in biaxially compressed ($\sigma^{(0)}=1$ GPa) Ge films on Si(001). The AE effect is maximum when the wave is still penetrating into the unstressed substrate (cf. Fig. 5). For [110] propagation the AE effect changes sign.

In Fig. 3 the calculated velocity change Δv is displayed for Rayleigh-type waves propagating parallel to the [100] and [110] direction when the Ge film is biaxially compressed by 1 GPa. For thick films and high frequency limit the velocity change approximates a constant value. But, in contrast to previous discussions, this is not the maximum velocity change of this system, which appears at $fh \simeq 700$ m/s. For the [100] direction the maximum of the AE effect is almost twice the value for a wave completely localized within the stressed film. For the [110] propagation direction the maximum of Δv is even opposite in sign with respect to the high frequency limit.

In order to explain this surprising phenomenon we separated the influence of the different mechanisms according to Eqs. (1)–(3) on the phase velocity: (i) the influence of residual stress on the equation of motion, (ii) the influence of residual strains on the modified stiffness tensor, and (iii) the influence of residual strains on density. For each scenario the phase velocity shift has been calculated separately for the [100] propagation and plotted in Fig. 4 as a function of the frequency-thickness product fh. The contribution of density and stress are different in magnitude; the strain contribution is even opposite in sign. Hence the total solution (solid line) exhibits a local maximum in the velocity change. For Ge/Si(001) at 1 GPa a maximum value of -20 m/s is observed at about $fh \simeq 700$ m/s.

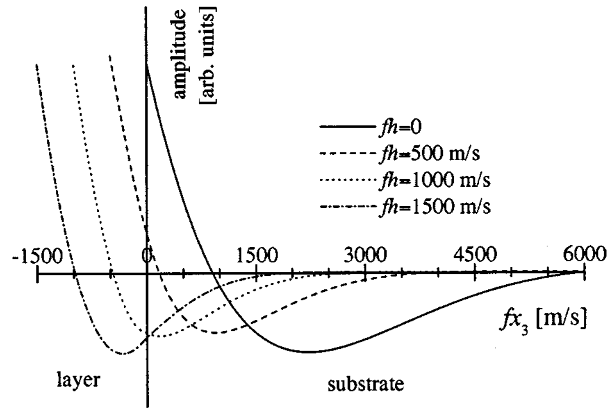

FIG. 5. Dependence of the amplitude of the longitudinal displacement component for the Rayleigh-type wave in the [100] direction vs the product of the frequency and penetration depth fx_3. The interface between the substrate and the layer corresponds to $fx_3=0$, while $fx_3<0$ and $fx_3>0$ represent the layer and substrate region, respectively.

In Fig. 5 the amplitude dependence of the longitudinal particle displacement component into substrate is shown for four fh values, starting at an infinitesimal film thickness (solid line). The amplitude of the longitudinal oscillation decreases rapidly when the wave penetrates into the substrate. For certain threshold values the oscillation phase changes its sign. When the layer thickness increases, the threshold approaches the interface. For $fh \gtrsim 700$ m/s the longitudinal oscillation with opposite phase enters the region of residual stress. This, however, causes a local velocity change in the

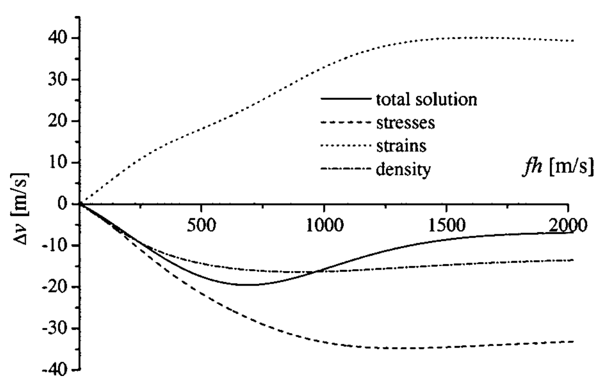

FIG. 4. Dependence of the velocity change Δv vs frequency-thickness product fh for Rayleigh-type waves propagating parallel to the [100] direction ($\sigma^{(0)}=1$ GPa) for the influence of (i) the residual stress on the equation of motion (dash); (ii) the residual strains on the modified stiffness tensor (dotted); and (iii) the residual strains on density (dash-dotted). The solid line represents the total solution shown in Fig. 3.

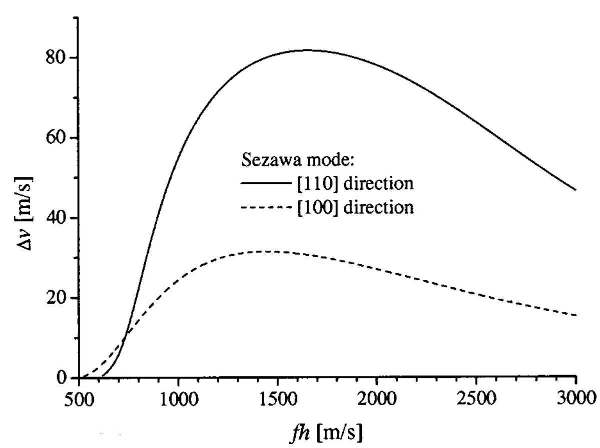

FIG. 6. Phase velocity change Δv vs frequency-thickness product fh for Sezawa waves propagating parallel to the [100] and [110] directions in biaxially compressed Ge films ($\sigma^{(0)}=1$ GPa) on Si(001). For the [110] direction the AE effect reaches its maximum of $\Delta v \simeq 82$ m/s at $fh=1660$ m/s.

opposite direction. Hence the integral AE effect is reduced. Consequently a maximum velocity change is observed (cf. Fig. 3).

In Fig. 6 the velocity change Δv vs fh of the Sezawa mode along [100] and [110] is plotted. Similar to the Rayleigh wave, again a maximum velocity shift occurs at finite film thickness. However, the AE effect of the Sezawa mode exceeds the value of the Rayleigh wave significantly. This behavior can be explained by the fact that in this case the longitudinal particle displacement component of the Sezawa wave exceeds the vertical component (in contrast to Rayleigh waves), thus leading to a considerably stronger AE effect. Along [110] a velocity change of 82 m/s is calculated for a Ge film biaxially compressed by 1 GPa, i.e., almost 1.7% with respect to the unstressed system. This enormous velocity change meets the accuracy of present ultrasonic devices. Therefore our results clearly demonstrate that the influence of stress on wave propagation has to be incorporated in current evaluation algorithms for the determination of elastic constants of heteroepitaxial thin films.

In conclusion we have investigated the acoustoelastic effect for biaxially stressed solid crystalline heterostructures. The change of the phase velocity of Rayleigh-type and Sezawa waves due to residual stresses reveal an anomalous frequency dispersion exhibiting a maximum at finite layer thickness, where the wave is still penetrating into the unstressed substrate. For Sezawa modes where the longitudinal oscillation component exceeds the vertical component we found a huge increase of the AE effect. Since the maximum AE effect can appear even for quite thin layers, i.e., when the layer thickness is much smaller then the acoustic wavelength, this has significant consequences for GHz acoustic phonon measurements, e.g., Brillouin spectroscopy and scanning probe microscopy, of highly stressed single crystalline heterostructures.

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