# Surface Brillouin scattering in semiconductor Fibonacci multilayers

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(Received 2 April 1990)

Brillouin spectra from surface (Rayleigh) acoustic waves in $a$-Si:H/$a$-SiN$_x$:H ($x \simeq 0.94$) Fibonacci multilayers have been measured with a tandem multipass Fabry-Pérot interferometer. To make a detailed analysis for the observed surface (Rayleigh) waves, following Merlin *et al.* [Phys. Rev. Lett. **55**, 1678 (1985)] and Dharma-wardana *et al.* [Phys. Rev. Lett. **58**, 1761 (1987)], we have formulated the propagation of surface acoustic waves parallel to the surface of the quasiperiodic multilayers using the Fourier-transform method in the long-wavelength regime. We found that the obtained Brillouin spectra can be accounted for by this new derivation of the Fourier transform of the surface acoustic waves, but a rather fascinating feature for the phonon frequency is that it is slightly modified by quasiperiodic indices $(n,m)$ or $p$. Such a modulation is attributed to the existence of a small but nonvanishing acoustic impedance. Our theoretical approach gives the correct order of amplitude modulation for surface (Rayleigh) waves within an error of $<1\%$. We thereby obtain an overall understanding of the elastic properties of the Fibonacci multilayers.

## I. INTRODUCTION

The study of surface acoustic modes in multilayers via Brillouin scattering is of great current interest due to the advent of high-contrast spectrometers.[1] The reason is that a number of novel materials can be prepared by artificially growing them in thin-film form, and such acoustic modes have taken on a new importance with regard to high-frequency acoustic-surface-wave devices for electronic signal processing. According to elastic theory, the surface-phonon spectra from Rayleigh, Sezawa, and Lamb waves depend considerably on the macroscopic physical parameters of the film, such as its density, elastic coefficients, and its thickness. Therefore, the behaviors of the surface acoustic waves are strongly affected by modifications that occur in thin films.

Although a number of experimental studies have been devoted to the hydrogenated amorphous silicon–silicon nitride systems,[2–5] where electrical as well as optical methods have been applied, the understanding of the elastic properties, especially for those of quasiperiodic structures, is still incomplete. Here we present data on Rayleigh surface-phonon spectra as well as the elastic properties of a quasiperiodic amorphous semiconductor multilayers via inelastic Brillouin scattering. These multilayers have a one-dimensional (1D) structure along the growth direction which is quasiperiodic; i.e., it is characterized by two different fundamental periods whose ratio is irrational. Such a quasiperiodic structure is of increasing interest because it is intermediate between the completely periodicity and randomness or disordered amorphous materials. On the other hand, the potential applications for multiband filters, integrated analogical frequency analyzer, and analogical coding, and so on, further motivate new theoretical and experimental studies.[6,7] Brillouin scattering has shown that is it a powerful nondestructive technique for studying surface acoustic waves supported by the films.[1]

In our work, as is the case for all Brillouin-scattering measurements from opaque materials, essentially only scattering from surface waves is observed. These waves propagate along the surface of the film and their amplitude decreases exponentially away from the surface. However, the appearance of the Sezawa and Lamb modes in the surface-phonon spectra depends on transverse sound velocity of the substrate ($v_t^S$) and that of the multilayer material ($v_t^M$). In other words, besides the Rayleigh phonon modes, the Sezawa or Lamb modes exist only if $v_t^S > v_t^M$. For simplicity, we have intentionally chosen glass slabs as substrate materials so that the surface (Rayleigh) waves are only observable. Simultaneously, the measurements of the surface (Rayleigh) waves could provide a critical test of our theoretical approach presented in Sec. III. Therefore, in this paper we only concerned ourselves with such surface acoustic waves, and relevant work in more complicated systems will be presented elsewhere.

To make a detailed analysis for the observed surface-phonon spectra, we further develop the effective-modulus model[8] based on the Fourier-transform method. A complete description of the Fourier transform for a quasiperiodic physical variation has been presented by Dharma-wardana *et al.* In Sec. III we will only outline the key points and then use the elastic continuum approximation to yield an explicit expression of the velocity of surface (Rayleigh) waves. Such a treatment to wave propagation in quasiperiodically layered systems is somewhat analogous to the invariant method presented by Sapriel *et al.*[9]

The structure of this paper is as follows. Section II describes the structures and characterizations of the Fibonacci multilayers. In Sec. III we present a theoretical treatment of surface (Rayleigh) waves polarized in the sagittal plane and derive the explicit expressions. Results from experiments and calculations are presented, and concluding remarks are made in Sec. IV. Section V is used for a brief summary and conclusions.

## II. SAMPLE CHARACTERIZATION OF FIBONACCI MULTILAYERS

The $a$-Si:H/$a$-SiN$_x$:H ($x \simeq 0.94$) quasiperiodic Fibonacci multilayers were prepared by a capacitive rf glow-discharge plasma technique and deposited onto a 7059 glass substrate, as described in Ref. 4. Pure silane was used to deposit the $a$-Si:H sublayers. The $a$-SiN$_x$:H sublayers were grown from a mixture of ammonia (eight parts) and silane (one part). A Fibonacci multilayer comprises an arrangement of layers of elements $A$ and $B$ following the Fibonacci sequence $S_1=A$, $S_2=AB$, $S_3=ABA,\ldots,S_j=\{S_{j-1}S_{j-2}\}$. A most remarkable characteristic feature of such a rule is self-similarity, and it is responsible for many of the spectral properties. The sequence $S_j$ consists of $F_j$ elements $A$ and $F_{j-1}$ elements $B$; $\{F_j\}$ is the $j$th term of the Fibonacci series defined iteratively by the recurrent law $F_j=F_{j-1}+F_{j-2}$, for $j\geq 2$, with $F_0=0$ and $F_1=1$. When $j$ increases, the rational number $F_j/F_{j-1}$ converges to the irrational golden mean $\tau=(1+\sqrt{5})/2$.

For the sample we studied here, each of the two basic elements $A$ and $B$ is composed of an $a$-Si:H well and an $a$-SiN$_x$:H barrier; namely, $A$ and $B$ were subdivided into an $a$-Si:H layer of thicknesses $d_1^A$ and $d_1^B$, respectively, adding an $a$-SiN$_x$:H layer of thickness $d_2$. Calculations based on the growth rate ($\sim 1.0$ Å/s) result in the following estimates of the layer thicknesses: $d_1^A=40$ Å, $d_1^B=17$ Å, and $d_2=20$ Å. A 12-generation multilayer was prepared according to the Fibonacci sequence and had a total thickness of 1.19 $\mu$m. Therefore, the approximate thicknesses of the $A$ and $B$ elements are $d_A=d_1^A+d_2=60$ Å and $d_B=d_1^B+d_2=37$ Å, respectively, and the quasiperiodicity is characterized by the $d=\tau d_A+d_B=134.1$ Å.

The quality of the sample is extremely important, since we are concerned with relatively small changes in the acoustic properties of the very thin multilayers. To determine the quality of our sample, we have completed the x-ray-diffraction measurements. The obtained diffraction patterns show that sharp peaks can be observed at low angles, as are expected for well-defined sublayers with different scattering factors. Moreover, the spacing between peaks corresponds to the quasiperiodicity $d$, and the scattering wave vector satisfies the relation of $k_{p,n}=k_{p-1,n}+k_{p+1,n}$, where $k_{p,n}=2\pi n\tau^p/d$. Meanwhile the high-resolution images suggest the interface mixing to be of the order of a few angstroms. Thus, from a structural standpoint, high-quality multilayers have been achieved.

## III. THEORETICAL BACKGROUND

To study the propagation of the sagittal-plane polarized surface acoustic waves in Fibonacci multilayers, a canonical procedure for calculating its dispersion is within the framework of the linear theory of elasticity to study the acoustic-wave equation[10]

$$\rho\frac{\partial^2 u_\alpha}{\partial t^2}=\sum_{\beta,\mu,\nu} C_{\alpha\beta\mu\nu}\frac{\partial^2 u_\mu}{\partial x_\beta \partial x_\mu},\quad \alpha=1,2,3 \tag{1}$$

where $u_\mu$ denotes the nonvanishing components of phonon displacement, and $C_{ijkl}$ is the elastic coefficients of the Fibonacci multilayers with thickness $h$ bounded by both stress-free surface and substrate. The stress-free boundary conditions are imposed at the air-multilayer interface, and the continuity of stress and displacement is applied at each interface and multilayer-substrate boundary as well. However, in what follows we shall use an alternative method, the Fourier-transform technique, to formulate the problem of the surface waves. The principal advantage of this method is that it can be used conveniently for periodic or quasiperiodic variations.

The multilayers can be considered as an elastic medium in the long-wavelength limit, and are due to the quasiperiodicity $d$ being much smaller than the wavelength $\lambda$ of the phonons. We introduce an orthonormal coordinate system $o$-$xyz$ with its $z$ axis along the quasiperiodic direction, $x$ and $y$ lying in the plane of the film. Because of the elastic isotropy of each medium, we can assume that each element has hexagonal symmetry with its axis of sixfold symmetry along the $z$ axis for simplicity. With no loss of generality, we assume wave propagation in the $y$ direction, whose displacement vector lies in the sagittal plane, which is only relevant to $C_{44}$, $\sigma_{23}$, and $\epsilon_{23}$. In what follows, $C_{\alpha\beta}^A$, $\sigma_{\alpha\beta}^A$, $\epsilon_{\alpha\beta}^A$ and $C_{\alpha\beta}^B$, $\sigma_{\alpha\beta}^B$, $\epsilon_{\alpha\beta}^B$ represent the components of the elastic constant, stress, and strain tensors, respectively. Superscripts $A$ and $B$ refer to the two elements, respectively, and $C_{\alpha\beta}^M$, $\sigma_{\alpha\beta}^M$, and $\epsilon_{\alpha\beta}^M$ refer to the effective properties of the multilayer. Let us start with the discussion of the transverse elastic waves polarized in the sagittal plane, which provides a relationship between the bulk transverse waves and the surface (Rayleigh) waves. In order to do this, we should derive first the structure factor of such a quasiperiodic sequence with use of the projection method[11] from a higher-dimensional periodic lattice. On the basis of a two-dimensional (2D) rectangular lattice, the whole set of reciprocal vertices labeled $(n,m)$ in reciprocal 2D space is projected onto a straight line whose direction is not crystallographic; one thereby gets a nonperiodic but quasiperiodic 1D sequence. This leads to a dense set of allowed 1D $k$ vectors, and the amplitude of the structure factor depends only on the indices $(n,m)$ and evidently on the contents of the two basic elements $A$ and $B$. From such an analysis, the structure factor associated with wave vector $k=k_{n,m}\equiv 2\pi d^{-1}(m+n\tau)$ can be expressed as[12,13]

$$S(k)=\frac{\tau^2}{d}\sum_{n,m}\frac{\delta_{k,k_{n,m}}}{2iX_{n,m}}(e^{2iX_{n,m}\tau^{-2}}-e^{-2iX_{n,m}\tau^{-1}}) , \quad (2)$$

where

$$X_{n,m}=\frac{\pi\tau[m(d_A/d_B)-n]}{\tau^{-1}+d_A/d_B} .$$

For an arbitrary function $\Phi(z)$ of position within each $A$ or $B$ element, with strain $\epsilon(z)$, effective sound velocity $v(z)$, and photoelastic coefficient $P(z)$, etc., its general form of the Fourier transform in the case of quasiperiodic structure is

$$\Phi(k)=S_A(k)\Phi_A(k)+S_B(k)\Phi_B(k) , \quad (3)$$

where $S_A$ and $S_B$ are substructure factors of two basic elements $A$ and $B$, and $\Phi_A$ and $\Phi_B$ describe the dependence of $\Phi$ on position within an element. The continuous use of the recurrent law gives the substructure factors to be

$$S_A(k)=\frac{\tau^2}{d}\sum_{n,m}\frac{\delta_{k,k_{n,m}}}{2iX_{n,m}}(e^{2iX_{n,m}\tau^{-2}}-e^{-2iX_{n,m}\tau^{-3}}) , \quad (4)$$

and

$$S_B(k)=\frac{\tau^2}{d}\sum_{n,m}\frac{\delta_{k,k_{n,m}}}{2iX_{n,m}}(e^{2iX_{n,m}\tau^{-4}}-e^{-2iX_{n,m}\tau^{-3}})e^{-ikd_A} . \quad (5)$$

According to the assumed symmetry of the elements, the stresses $\sigma_{13}^i$, $\sigma_{23}^i$, $\sigma_{33}^i$ and the strains $\epsilon_{11}^i$, $\epsilon_{12}^i$, as well as $\epsilon_{22}^i$, can be treated as invariants, where $i=A$ and $B$. Note that it may be not adequate for evaluating $\sigma_{11}^M$, $\sigma_{12}^M$, $\sigma_{22}^M$, $\epsilon_{13}^M$, $\epsilon_{23}^M$, and $\epsilon_{33}^M$ using the constituent average for a quasiperiodic structure.[8,9] After having taken into account this point, based on Eq. (3), we can write down the strain expression in reciprocal space

$$\epsilon_{23}^M(k)=S_A(k)\epsilon_{23}^A(k)+S_B(k)\epsilon_{23}^B(k) , \quad (6)$$

where $\epsilon_{23}^A(k)$ and $\epsilon_{23}^B(k)$ are the Fourier transforms of the strains $\epsilon_{23}^A(z)$ and $\epsilon_{23}^B(z)$ within $A$ and $B$ elements, respectively. Here the strain $\epsilon_{23}^i(k)$ can be readily yielded by the following general expression:

$$F_j(k)=\int_0^{d_j}e^{-ik\xi}F_j(\xi)d\xi, \quad j=A \text{ or } B \quad (7)$$

where $F_j(\xi)$ describes the dependence of $F$ on position within a $j$ element. Therefore, we can find that $\epsilon_{23}^A(k)=\epsilon_{23}^A[1-\exp(-ikd_A)]/ik$ and $\epsilon_{23}^B(k)=\epsilon_{23}^B[1-\exp(-ikd_B)]/ik$; meanwhile, with respect to the stress $\sigma_{23}$, the associated Fourier transform of the strain in Eq. (6) is reduced to

$$\epsilon_{23}^M(k)=\bar{\epsilon}_{23}\delta(k)+(\epsilon_{23}^A-\epsilon_{23}^B)\sum_{n,m}\delta(k-k_{n,m})S(k_{n,m}) , \quad (8)$$

where the zeroth-order term

$$\bar{\epsilon}_{23}=(\epsilon_{23}^A\tau d_A+\epsilon_{23}^B d_B)/d , \quad (9)$$

which is identical to the usual effective-modulus model of the constituent average. The second term in Eq. (8) is clearly meant to represent a corrective one to the effective-modulus model for the quasiperiodic case as being due to the existence of a small but nonvanishing relative strain in the interfaces of the multilayers. The $\delta$ function $\delta(k-k_{n,m})$ determines the quasiperiodic modulation of the strain only occurring at the wave vector $k\equiv 2\pi(m+n\tau)/d$, and at the same time, its modulation amplitude is dependent on the structure factor $S(k_{n,m})$ of the quasiperiodic lattice. If we do not concern ourselves very much with this term, i.e., consider only the contribution from the first term that corresponds to $n=m=0$, and then the velocity of the surface (Rayleigh) waves of the zeroth-order approximation can be derived according to the procedures below.

By first applying Hook's law to each basic element ($A$ and $B$), the effective medium leads to

$$\epsilon_{23}^i=\sigma_{23}^i/C_{44}^i, \quad i=A,B,M . \quad (10)$$

However, the stress $\sigma_{23}$ within each basic element should satisfy the condition of $\sigma_{23}^M=\sigma_{23}^A=\sigma_{23}^B$, which actually arises from the continuity of the normal components of the stress at each interface. On the other hand, using similar procedures, the density $\rho(z)$ can also be expressed in a Fourier series:

$$\rho^M=\bar{\rho}\delta(k)+(\rho_A-\rho_B)\sum_{n,m}\delta(k-k_{n,m})S(k_{n,m}) , \quad (11)$$

and the first term refers to the spatial average

$$\bar{\rho}=\frac{(\tau d_A\rho_A+d_B\rho_B)}{d} , \quad (12)$$

where $\rho_A$ and $\rho_B$ are the mass densities of the two basic elements, respectively.

Generally speaking, the transverse elastic wave velocity in a quasiperiodic amorphous structure in the form of $v_t^M=(C_{44}^M/\rho^M)^{1/2}$ seems to be adequate, because in a nonpathological system the modulus of the Fourier components $\rho_i$ and $\epsilon_i$ is a rapidly decreasing function of $i$. On the other hand, since the relative density $|\rho_A-\rho_B|$ and the relative strain $|\epsilon_{23}^A-\epsilon_{23}^B|$ are much smaller than the corresponding relative differences $|\rho_{a\text{-Si:H}}-\rho_{a\text{-SiN}_x\text{:H}}|$ and $|\epsilon_{23}^{a\text{-Si:H}}-\epsilon_{23}^{a\text{-SiN}_x\text{:H}}|$, respectively, we may keep only the zeroth-order term of both the density $\rho^M$ and the strain $\epsilon_{23}^M$. Therefore, the most drastic approximation consists in retaining only $\bar{\epsilon}_{23}$ and $\bar{\rho}$. When Eqs. (10) and (12) are substituted into Eq. (9), after some algebraic treatments, the velocity of the transverse acoustic waves in the sagittal plane can then be expressed as

$$v_t^M=d\left[\frac{\tau^2 d_A^2}{C_{44}^A/\rho_A}+\frac{d_B^2}{C_{44}^B/\rho_B}+\frac{\tau d_A d_B}{(C_{44}^A/\rho_A)^{1/2}(C_{44}^B/\rho_B)^{1/2}}\left[\frac{\rho_A(C_{44}^A/\rho_A)^{1/2}}{\rho_B(C_{44}^B/\rho_B)^{1/2}}+\frac{\rho_B(C_{44}^B/\rho_B)^{1/2}}{\rho_A(C_{44}^A/\rho_A)^{1/2}}\right]\right]^{-1/2} . \quad (13)$$

In the above equation, the ratio $[\rho_A(C_{44}^A/\rho_A)^{1/2}]/[\rho_B(C_{44}^B/\rho_B)^{1/2}]$ reflects the size of the acoustic impedances of the two constituting elements; while $(C_{44}^i/\rho_i)^{1/2}$ is the bulk transverse velocity within the corresponding $i$ elements.

Since the surface (Rayleigh) waves in an isotropic free-surface propagation can be approximated by the explicit equation $v_R=\beta v_t$, with $\beta=(0.87C_{11}+2C_{12})/(C_{11}+2C_{12})$. For the cylindrical symmetry, the associated velocity of surface (Rayleigh) waves should have the identical form, but the $\beta$ only involves a linear combination of $C_{13}$, $C_{11}$, and $C_{33}$. For amorphous multilayers, owing to the arbitrary choice of the contents of the two elements, as has been done in our case, the $\beta$ can be approximated by $\beta^M \simeq \beta^A \simeq \beta^B$. This approximation has been found to be a reasonable one for the periodic $a$-Si:H/$a$-$SiN_x$:H superlattice case.[14] Based on such a consideration it follows that $v_R^M=\beta^M v_t^M$, and the corresponding analytical solution is

$$v_R^M=d\left[\left[\frac{\tau d_A}{v_R^A}\right]^2+\left[\frac{d_B}{v_R^B}\right]^2+\frac{\tau d_A d_B}{v_R^A v_R^B}\left[\frac{\rho_A v_R^A}{\rho_B v_R^B}+\frac{\rho_B v_R^B}{\rho_A v_R^A}\right]\right]^{-1/2}, \quad (14)$$

where $v_R^i$ for $i=A$ and $B$ is the corresponding velocity of surface (Rayleigh) waves propagating parallel to the surface of the two basic elements. Note that this velocity of the surface (Rayleigh) waves differs slightly from the one obtained from the effective-modulus model.[15]

Furthermore, to understand the influence of the corrective term, one can directly discuss it through perturbation theory, as has already been done in the case of periodic multilayers.[16] We would like, however, to continue the discussion using the Fourier-transform method, starting with Eqs. (3)–(5). Here the arbitrary function $\Phi(z)$ represents the velocity $v_R(z)$ of the surface (Rayleigh) waves, propagating parallel to the surface of the multilayers. By assuming that the velocities of surface (Rayleigh) waves corresponding to the two basic elements are, respectively, $v_R^A$ and $v_R^B$, ignoring detailed calculations, we can almost immediately write the Fourier transform of $v_R(z)$ in a similar form as in Eqs. (8) and (11). However, it should be pointed out that since $n$ and $m$ span all integers, the number of nonzero Fourier components in an arbitrary interval is infinitely dense, so that only the greatest Fourier components would modify the surface acoustic waves. These components only occur in the subset of $k_{n,m}$, where $n$ and $m$ are neighboring Fibonacci numbers, and hence $k_{n,m}=2\pi(F_{p-1}\tau+F_p)d^{-1}=2\pi\tau^p d^{-1}$, where $p$ is an integer. After having replaced the quasiperiodic indices $(n,m)$ by $p$, we can easily verify that the following equations should hold:

$$\bar{v}_R(0)\simeq v_R^M=\beta^M v_t^M, \quad n=m=0\ , \quad (15)$$

$$v_R\simeq\bar{v}_R(0)+\frac{d}{\pi^2 d_A}(v_R^A-v_R^B)\times\sum_p\left|\sin\left[\frac{\pi d_A\tau^{1-p}}{d}\right]\sin\left[\frac{\pi d_A\tau^p}{d}\right]\right|, \quad (16)$$

where $v_R$ is the velocity of the surface (Rayleigh) waves in the Fibonacci multilayers with $d_A/d_B=\tau$, and $p$ is any integer, whether positive, negative, or zero. In doing the above equations, we have taken advantage of the existence of self-similarity symmetry in quasiperiodic structures, and, have chosen the origin of the coordinates to be in the middle of an arbitrary triplet structure to obtain real Fourier components. As can be seen from Eq. (16), the modulation amplitude of the velocity of surface (Rayleigh) waves around $\bar{v}_R(0)$ amounts to

$$\Delta\simeq\sum_p v_R^{(p)}=\frac{d}{\pi^2 d_A}(v_R^A-v_R^B)\sum_p\left|\sin\left[\frac{\pi d_A\tau^{1-p}}{d}\right]\sin\left[\frac{\pi d_A\tau^p}{d}\right]\right|\ . \quad (17)$$

Obviously, when the relative difference between $v_R^A$ and $v_R^B$ is not ignored, such a quasiperiodic modulation will in fact lead to a larger deviation of the measured $v_R$ compared to predicted data in terms of Eq. (14). In contrast, for a much smaller relative difference between $v_R^A$ and $v_R^B$, the associated quasiperiodic modulation is generally weak compared with $\bar{v}_R(0)$, and Eq. (16) could be therefore reduced to Eq. (14), the solution of the zeroth-order approximation.

## IV. RESULTS AND DISCUSSION

The Brillouin-scattering experiments were performed in backscattering geometry using a high-contrast (3+3)-pass tandem Fabry-Pérot interferometer, operating at a free spectral range of about 45 GHz. A single-mode 5145-Å $Ar^+$-ion laser was used with an incident power of $p$-polarized light of up to 60 mW. The sampling time per spectrum was typically 1.5 h. The polarization of the scattered light was not analyzed in order to achieve a reasonably large scattering signal. An acousto-optic modulator was used to attenuate the light when scattering through the intense light peaks at the laser frequency. The dispersion of the Rayleigh mode was measured by varying $\theta_i$, the angle of incidence, and hence $q$ ($2\kappa_i\sin\theta_i$, where $\kappa_i$ is the wave vector of the incident beam), the in-plane phonon wave-vector component. Here only $q$ is conserved, so that for the backscattering configurations, the scattered photons are frequency shifted by the acoustic frequency $\omega$. By dividing $q$, the surface wave velocity $v_R=\omega/q$ is able to be determined by the experiments. All measurements were made in air at room temperature.

### A. Surface-phonon spectra

Typical Brillouin spectra from several incident angles measured at $\theta_i=75°$, 65°, and 45° are shown in Fig. 1 and display increasing linewidth with decreasing incident angles. Owing to the intentional choice of substrate material, the observed inelastic spectral lines symmetrical around the central peak should be associated with the surface (Rayleigh) phonon modes, whose frequency strongly depends on the incident angle $\theta_i$, defined by the

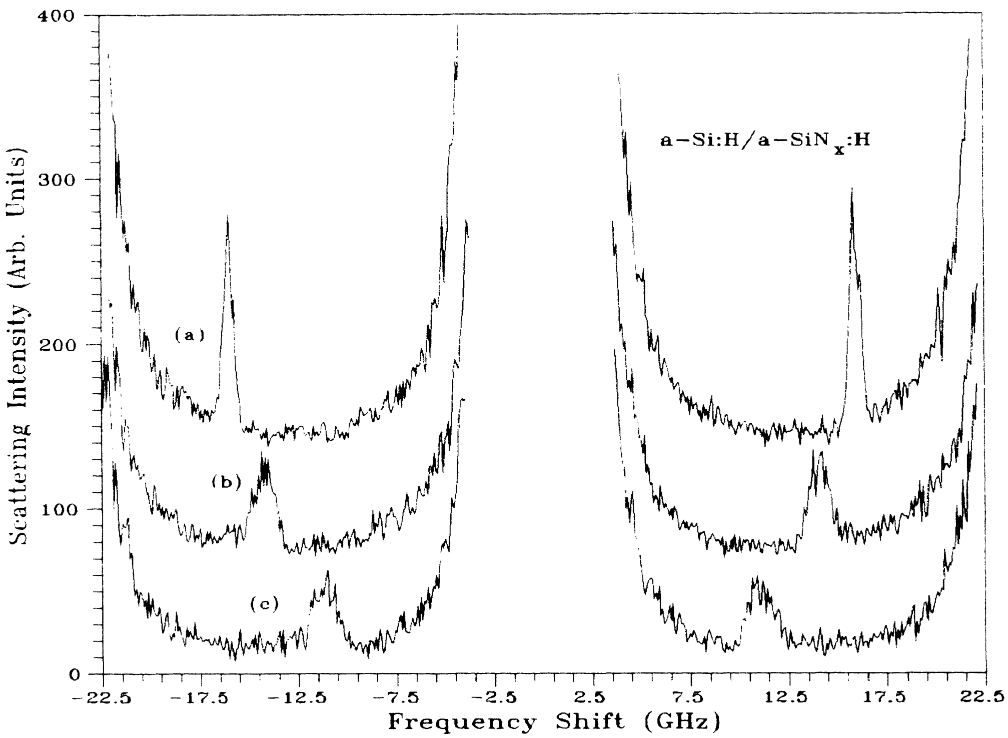


FIG. 1. Brillouin spectra of Fibonacci $a$-Si:H/$a$-SiN$_x$:H multilayers consisting of 12 generations with a quasiperiodicity of $d = 134.1$ Å: (a) $\theta_i = 75°$, (b) $\theta_i = 65°$, and (c) $\theta_i = 45°$. Note that the ghost peaks appear only greater than 18 GHz due to elastic scattering.

angle between the incoming light beam and the surface normal. However, bulk acoustic modes were not observed due to the limitations of the smaller free spectral range (45 GHz), and due to the opacity of the $a$-Si:H/$a$-SiN$_x$:H multilayers. In particular, a splitting of the Brillouin peaks was obviously observed for all scattered angles. This evolution of the line shapes with the incident angle can be simulated by the wave-vector dependence of the scattering cross section. Of course, if the multilayer is not thick enough, the Brillouin spectrum from the substrate will be superposed to that of the multilayer. For our case, this influence can be neglected, because the penetration depths of the incident light in $a$-Si:H/$a$-SiN$_x$:H are much smaller than the total thickness of the Fibonacci multilayers. With respect to measurements of the peak dispersion, we have determined the velocity of surface (Rayleigh) waves to have the value of $v_R = 4.10 \times 10^5$ cm/s. In Fig. 1 the central and ghost peaks are due to the elastic scattering.

A further check on the surface (Rayleigh) waves was carried out by measuring the surface-phonon spectra corresponding to the $a$-Si:H and $a$-SiN$_x$:H sublayers under the same experimental conditions. We obtained $v_R$($a$-Si:H)$= 4.18 \times 10^5$ cm/s; but the results of $v_R$($a$-SiN$_x$:H)$= (3.80 \pm 0.06) \times 10^5$ cm/s are only evaluated from our previous ultrasonic measurements.[4,15] This is because the $a$-SiN$_x$:H film is an insulator whose smaller dielectric constant makes the coupling between light and the surface modes by the ripple mechanism very weak and consequently of a low scattering efficiency.[17] Whereas, the velocity of surface (Rayleigh) waves can be generally expressed as a function of the dimensionless quantity $qh$. With increasing $qh$, especially for $qh \to \infty$, the velocity of the Rayleigh waves converges rapidly to the Rayleigh velocity of the multilayers; in contrast, its velocity almost approaches the value of the substrate material in the limit of $qh \to 0$. In the case of $qh \sim 20$, as in our sample, the observed $v_R$ should be the value of the Fibonacci multilayers. We are therefore convinced that the observed surface-phonon spectra are associated with the surface (Rayleigh) waves of the Fibonacci multilayers.

### B. Applicability of the effective-modulus model

In what follows we first discuss the applicability of the effective-modulus model to $a$-Si:H/$a$-SiN$_x$:H Fibonacci multilayers with regard to the zeroth-order approximation. Such an approximation is to keep only the first term of the Fourier transform of the $v_R$, ignoring the corrective effects from higher-order terms. The approximation regards the Fibonacci multilayers as an effective medium, as we have discussed in Sec. III. From Eq. (14) one can find that the evaluation of the velocity $v_R^M$ of surface waves actually depends on $v_R^A$, $v_R^B$, and the ratio of $\rho_A/\rho_B$, respectively. According to the works of Santos *et al.*[3] and Liu *et al.*,[18] the ratio of the acoustic impedances of $a$-Si:H and $a$-SiN$_x$:H, $F = \rho_1 v_1 / \rho_2 v_2$, equals 1.4, and $v_2/v_1 = 0.74$, where subscripts 1 and 2 denote $a$-

Si:H and $a$-SiN$_x$:H sublayers, respectively. It follows that the density ratio $\rho_1/\rho_2 \simeq 0.56$. Mass densities $\rho_A$ and $\rho_B$ are well described by the following equations:

$$\rho_A = \frac{\rho_1 d_1^A + \rho_2 d_2}{d_1^A + d_2}, \quad \rho_B = \frac{\rho_1 d_1^B + \rho_2 d_2}{d_1^B + d_2}, \tag{18}$$

which determine the ratio $\rho_A/\rho_B \simeq 0.88$. On the other hand, a Fibonacci lattice can be regarded as a superposition of two periodic elements of period proportional to $F_j$ and $F_{j-1}$, so that $v_R^A$ and $v_R^B$ are then yielded by

$$v_R^i \simeq d_i \left[ \left[ \frac{d_1^i}{v_R^1} \right]^2 + \left[ \frac{d_2}{v_R^2} \right]^2 \right]^{-1/2}, \quad i = A,B \tag{19}$$

where $v_R^1$ and $v_R^2$ are the velocities of the surface waves in the $a$-Si:H and $a$-SiN$_x$:H sublayers. In this approximation, using the measured values of $v_R^1$ and $v_R^2$, the corresponding velocities of surface waves within elements $A$ and $B$ are, therefore $(4.05 \pm 0.02) \times 10^5$ cm/s and $(3.97 \pm 0.02) \times 10^5$ cm/s, respectively. Accordingly, the velocity $v_R^M$ of the zeroth-order approximation is equal to $(4.02 \pm 0.02) \times 10^5$ cm/s. This result seems to be comparable to the experimental value $4.10 \times 10^5$ cm/s to some extent, whereas an error of $\sim 2\%$ actually exists. A detailed analysis of the error, i.e., for basic element densities and sublayers thickness changes, etc., has ruled these out as possible causes.

### C. Quasiperiodic modulation

In order to identify this error in terms of the acoustic properties of the Fibonacci multilayers, we associate it with the quasiperiodic modulation of the surface acoustic waves. From previous results, we can calculate that the relative difference $v_R^A - v_R^B$ is about $0.08 \times 10^5$ cm/s. Although this value is smaller than $v_R^A$ or $v_R^B$, the effect of the second term in Eq. (16) on $v_R$ may be not negligible. Through calculations, we found that although the quasiperiodic index $p$ spans all integers, the primary modulation on $v_R$ is only associated with the few largest Fourier components. As can be seen in the structure-factor formula and in Fig. 2, the dominant components correspond to $|p| \leq 10$. Moreover, analogous to Raman spectra of quasiperiodic (Fibonacci) superlattices, the feature of self-similarity is able to be observed simultaneously.[12,19,21,22]

Figure 3 shows experimental results and theoretical fitting curves. The effective-modulus model approximately gives a linear relation between the velocity of surface waves and thickness ratio $\gamma$, defined by $\tau d_A/d$. The contributions from the quasiperiodic modulation term, however, give rise to a larger $v_R$ than $v_R(0)$. The measured results have been denoted by a dot and with an error bar. Obviously, within the error bar our obtained results are in good agreement with theoretical predictions. For $\gamma = 0.725$, the corresponding $v_R$ is $(4.13 \pm 0.02) \times 10^5$ cm/s. In this case, we find it comparable to the experimental value of $4.10 \times 10^5$ cm/s within an error of 0.7%. In Fig. 3 we also have indicated that the greatest quasiperiodic modulation of the surface waves, shown by an arrow, is less or equal to the velocity of surface waves of crystalline silicon for a smaller thickness ratio.

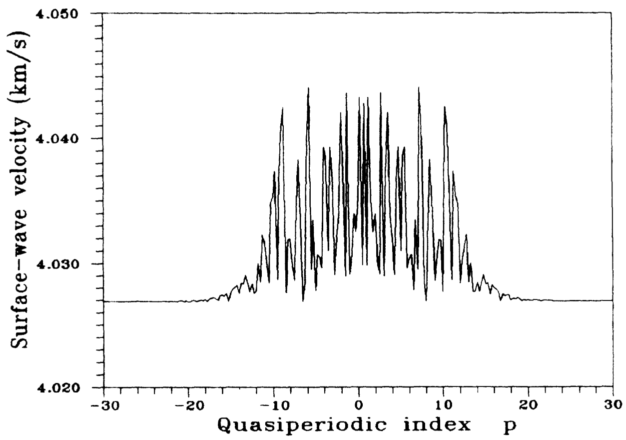


FIG. 2. Surface-wave velocity vs quasiperiodic modulation. The most prominent modulations only occur for $|p| \leq 10$ and can be characterized by the self-similarity symmetry.

In Eq. (17), an interesting conclusion is obvious: if the relative difference $v_R^A - v_R^B$ of the surface (Rayleigh) waves with regard to two basic elements is less than zero, the quasiperiodic modulation will then lead to a negative correction on the velocity $v_R(0)$ of surface waves, and the associated so-called phonon softening or elastic anomalous phenomenon[14,20] may be observed. In fact, the choice of the basic elements $A$ and $B$ is intentional, so that the observed results deviating from the effective-modulus model should be well expected. Such a conclusion suggests that the elastic anomalous may be a general phenomenon, which exists not only in periodic superlattices but also probably in quasiperiodic multilayers or superlattices as well. We hope this kind of prediction will be seen in further Brillouin-scattering experiments.

## V. CONCLUSIONS

In this paper we have shown that the acoustic properties of Fibonacci multilayers can be characterized by sur-

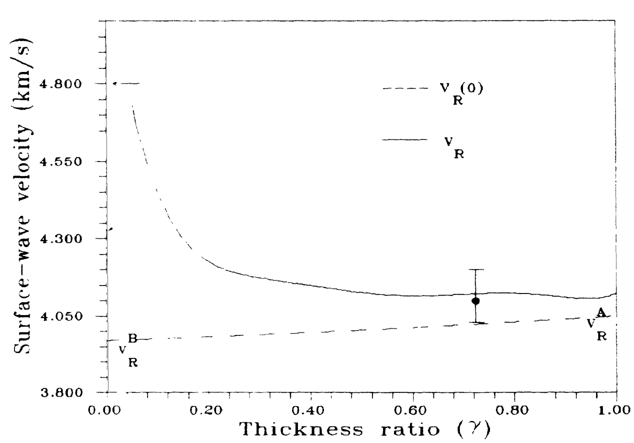


FIG. 3. Comparison of the surface-wave velocities calculated with the correction term (solid curve) and without correction (dashed line) based on the elastic continuum approximation. The dot represents the experimental results with an error bar.

face Brillouin-scattering experiments. The measured velocity of surface waves of the Fibonacci ($a$-Si:H/$a$-$SiN_x$:H) multilayers is well described by our theoretical approach based on the Fourier-transform method. The quasiperiodic modulation of the velocity of the surface waves is mainly dependent on the relative difference $|v_R^A - v_R^B|$ of the velocity of surface waves between two basic elements. In the first case, when this relative difference is not negligible, we found that such a modulation can be characterized by a well-known feature of self-similarity, and is somewhat analogous to the hierarchical structure of the Raman spectra.[12,19,21,22] The significant modulation of the quasiperiodic structure on the Brillouin spectra primarily arises from several lower quasiperiodic indices or from the few largest Fourier components. In contrast, when this relative difference is much smaller, which is associated with a vanishingly small acoustic impedance existing in the multilayers, it will give rise to a negligible correction of the effective-modulus model. Our discussions only focused on the specific cases of $d_A/d_B = \tau$ and surface (Rayleigh) waves, but our theoretical treatments and experimental studies are able to be extended to more general systems. For arbitrary $d_A/d_B \neq \tau$, a straightforward Fourier transform of transverse waves with the sagittal-plane polarized may give an explicit expression of the surface (Rayleigh) waves. However, the formulation of higher-order surface waves is more complicated because these surface waves are not only associated with the properties of both the multilayers and substrate materials, but also relate to those of the interface structures between the multilayer and substrate as well. Therefore, further theoretical studies are highly desirable for surface Brillouin scattering in quasiperiodic layered structures.

## ACKNOWLEDGMENTS

We would like to acknowledge Dr. G.M. Mao and Dr. Z. F. Li for help in sample preparations. This research was supported by the National Natural Science Foundation of China.

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