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Phonon stop bands in amorphous superlattices

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In periodically layered media the phonon dispersion relation shows energy ranges in which phonon propagation is not possible. The existence of such phonon stop bands in crystalline superlattices has been observed in work by V. Narayanamurti, H. L. Störmer, M. A. Chin, A. C. Gossard, and W. Wiesman [Phys. Rev. Lett. **43**, 2012 (1979)]. In this Rapid Communication we report the observation of phonon stop bands in amorphous superlattices. The filter characteristic of these amorphous superlattices is much sharper than in the case of the crystalline superlattices studied earlier. The investigated superlattices have been prepared by alternating evaporation of Si and SiO_2 layers as well as by plasma-enhanced chemical vapor deposition of a-Si:H/a-SiN_x:H films in a glow-discharge reactor.

I. INTRODUCTION

The recent development of thin-film technology has made it possible to prepare periodic multilayer structures consisting of an alternating sequence of crystalline or amorphous films of nanometer thickness. The periodic structure of such multilayers is the origin of some new properties. One of these properties is the "backfolded" phonon dispersion relation along the direction perpendicular to the superlattice. This backfolding is a result of the periodic modulation of the density and the elastic constants in the superlattice and it leads to gaps in the phonon energy at the center and the boundaries of the new Brillouin zone. Excitation of the "folded" acoustic phonons can be observed by Raman scattering.[1] In this case coupling mostly is limited to longitudinal phonons.

The phonon transmission characteristic of superlattices can be investigated by using phonon absorption spectroscopy methods. Phonon spectroscopy with superconducting tunnel junctions provides information about longitudinal and transverse acoustic phonon propagation. Because of the higher density of phonon states, transverse phonons are mostly predominant. For a detailed presentation of the methods used in phonon spectroscopy measurements we refer the reader to Ref. 2.

Corresponding measurements with superconducting tunnel junctions as generators and detectors of high-frequency acoustic phonons were first performed on crystalline GaAs/$Al_xGa_{1-x}As$ superlattices prepared by molecular-beam-epitaxy (MBE) techniques.[3] In the present Rapid Communication phonon absorption spectroscopy is used to investigate the phonon "filter characteristic" of amorphous Si/SiO_2 and Si/SiN_x superlattices.

II. SAMPLE PREPARATION

The Si/SiO_2 samples were fabricated in an automatic ultrahigh-vacuum (UHV) evaporation system. The starting pressure for all process steps was below 10^{-8} mbar. The composition of the residual gas could be examined using a quadrupole mass spectrometer. An electron gun was used to evaporate the deposited material. Alternating layers of SiO_2 and Si were deposited on one side of a Si crystal. During evaporation a quartz-crystal monitor was used to control the film thickness and growth rates. After the deposition of the multilayer an Al-Ox-Al tunnel junction was prepared on top of the superlattice and a Sn-Ox-Sn junction on the opposite crystal surface. The preparation of the tunnel junctions could be done without breaking the vacuum.

The Si/SiN_x superlattices were prepared by a plasma-enhanced chemical vapor deposition technique in a capacitive rf glow-discharge reactor. The deposition temperature was $T=280\,^{\circ}C$. A more detailed description of preparation conditions is given in Ref. 1. The individual thicknesses of the single layers were adjusted by the deposition time, using the known growth rate of each material.

III. PHONON SPECTROSCOPIC MEASUREMENT ON Si/SiO_2 SUPERLATTICES

Figure 1 shows the phonon transmission spectrum of a Si/SiO_2 multilayer structure with seven periods. In Fig. 1(a) phonons were generated in the Sn tunnel junction. They propagated through the substrate and the superlattice and were detected by an Al junction on top of the superlattice. In this arrangement the useful frequency range is from 130–285 GHz. In the upper curve of Fig. 1(a) the phonon generator was covered with oil to suppress additional structures in the investigated frequency range caused by a preferred phonon transmission from the generator to the surrounding He bath. In Fig. 1(b) the Al junction was used as phonon generator and the Sn junction as phonon detector. The useful frequency range in this case extends from 285 GHz up to 3 THz. The special arrangement of the experiment is demonstrated in the in-

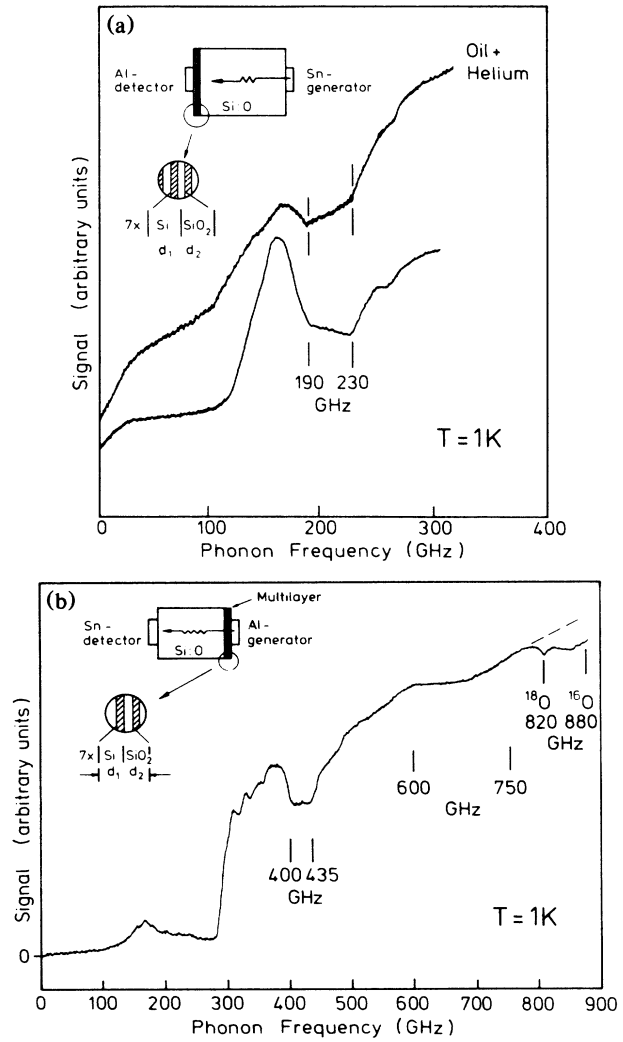

FIG. 1. Phonon transmission spectrum of an amorphous Si/SiO_2 superlattice.

sets of each figure.

The spectra exhibit strong and broad absorption bands with center frequencies around 210 and 420 GHz. A third but much weaker structure appears between 600 and 750 GHz. At 880 and 820 GHz additional absorption structures caused by the resonance of interstitial oxygen atoms in the Si substrate crystal can be recognized. The contribution of oxygen in the amorphous Si layers to this resonance structure is still an open question today. The demonstrated measurements give strong evidence for phonon stop bands in the evaporated Si/SiO_2 multilayer structures. In order to verify this interpretation the measurements may be compared with calculations of the phonon transmission coefficient based on a formalism known from classical optics.[4] The analogy is made by substituting for the impedance of the electromagnetic wave, the acoustic impedance $Z_i = \rho_i V_i$ of the individual layers with density ρ_i and sound velocity V_i. In this way the specific properties of the Si bulk crystal and of the tunnel junctions are not included in this calculation. Figure 2 shows a corresponding calculation to be compared with the measurements of Fig. 1.

In this calculation we have used the value of 3330 m/s for the transverse sound velocity in a-SiO_2, as obtained from phonon spectroscopy[5] for single SiO_2 layers evaporated under the same conditions as ours. Since this value differs from the value of fused SiO_2 by 10%, we assumed the same reduction for a-Si compared to the value of bulk Si. Since only the ratio of the density of both materials enters the calculation we have used the bulk values, assuming that any influence of the preparation technique is the same for both films. Because of the difficulties in measuring the absolute thicknesses of every single film during the evaporation process, the thicknesses of the Si and SiO_2 single films were used as fitting parameters. The best fit was achieved with $d_{Si} = 70$ Å and $d_{SiO_2} = 30$ Å. This is in reasonable agreement with the values measured by a quartz-crystal film-thickness monitor of $d_{Si} = 40$ Å and $d_{SiO_2} = 40$ Å, respectively.

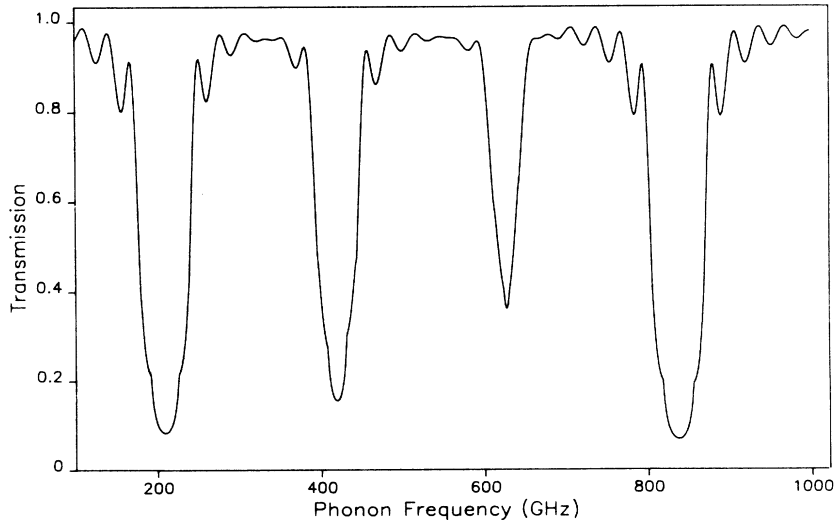

FIG. 2. Calculated phonon transmission for the superlattice of Fig. 1 with the following parameters: Si: $d_1 = 70$ Å, $v_1 = 4700$ m/s, and $\rho_1 = 2.2$ g/cm^3; SiO_2: $d_2 = 30$ Å, $v_2 = 3300$ m/s, and $\rho_2 = 2.3$ g/cm^3.

A comparison of the experimental results with the calculated transmission characteristic gives evidence that the small well-resolved structures outside the real stop bands can be associated with the side lobes of the stop-band characteristic. In the frequency range above 600 GHz the measured stop-band characteristic is significantly wider and weaker than expected from the calculations. This can be possibly attributed to phonon scattering in the SiO_2 layers, which is expected also from thermal conductivity measurements. Another possible reason is that with decreasing phonon wavelength the model of uniform, pinhole free layers with well-defined boundaries between the films no longer holds.

Looking at the absorption lines originating from the interstitial oxygen atom in the substrate, we see that even the small satellite of the ^{18}O isotope is well resolved. This indicates that high-frequency phonons outside the stop bands can propagate through many amorphous films of about 700-Å total thickness without or with only little reduction. It is further remarkable that the observation of the oxygen line gives evidence also for the high acoustic quality of the Si/SiO_2 surfaces, which do not produce strong scattering either. For justification of the reliability of the fabrication process we reduced the film thickness of each layer by a factor of 1.7. The respective phonon absorption spectrum is shown in Fig. 3.

Corresponding calculations indicate that a decrease of layer thickness leads to an increase of the center frequency of the stop bands which scales with the same factor. Therefore, frequency shifts of the first stop band from 210 to 360 GHz, and of the second stop band from 420 to 720 GHz are predicted. These expectations are fully confirmed in Fig. 3. Only the second resonance is widened in the same way as observed in Fig. 1(b) and additionally broadened by the interference with a phonon resonance in the very thin film of the Al generator junction.

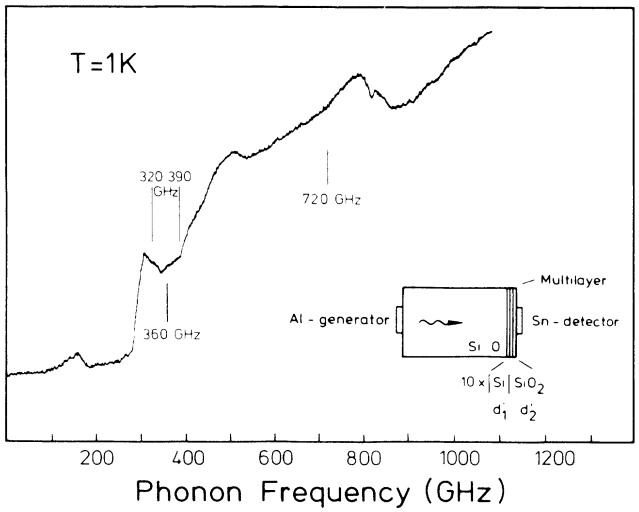

FIG. 3. Phonon transmission spectrum of a superlattice with film thickness reduced by a factor of 1.7 compared to those of Fig. 1.

IV. PHONON SPECTROSCOPY MEASUREMENTS ON *a*-Si:H/*a*-SiN_x:H SUPERLATTICES

In addition to Raman measurements, phonon spectroscopy measurements were also applied to *a*-Si:H/*a*-SiN_x:H superlattices.[6,7] Figure 4 shows the phonon transmission and backscattering spectrum of a multilayer structure consisting of 130 periods with an individual period thickness of 90 Å. The layer thicknesses were determined as described in Sec. II.

In the phonon transmission spectrum the phonons were detected by a Sn tunnel junction on top of the multilayer and generated by an Al junction at the opposite face of the substrate. In the phonon backscattering spectrum the phonon generator and detector were evaporated on the same crystal surface. The detailed arrangement of the measurement is shown in the inset of Fig. 4.

The phonon transmission spectrum exhibits a strong absorption band centered at 570 GHz. Another absorption band coincides with the detector threshold at 285 GHz. The 2:1 ratio of the center frequencies of these absorption bands gives strong evidence for identification of these structures as the first and second stop bands of the TA phonons. This is in good agreement with Raman measurements performed on the same sample.[7] The third stop band expected at 860 GHz is widened in the same way known from Si/SiO_2 multilayers. Moreover, it is additionally superposed on the resonance of oxygen in silicon. The main absorption bands are accompanied by side structures at 365 and 730 GHz which can be interpreted as interior band gaps due to phonons with propagation directions oblique to the interfaces, as will be discussed in a separate paper.[7] Related theoretical results were obtained by Tamura and Wolfe.[8]

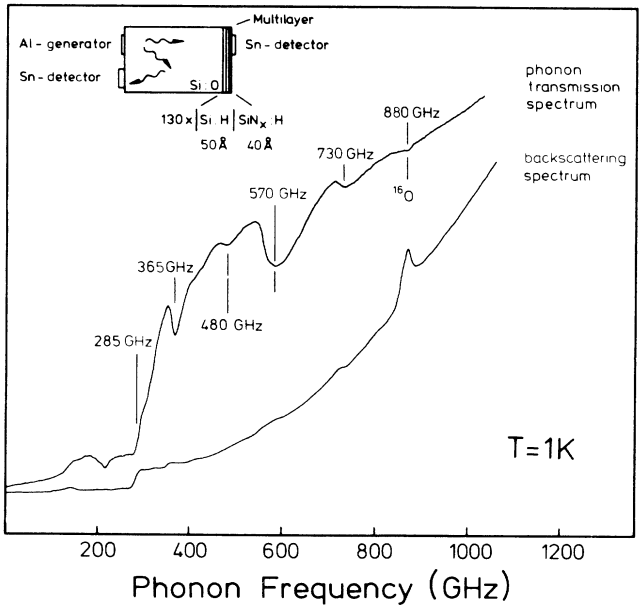

FIG. 4. Phonon transmission and backscattering spectrum of an *a*-Si:H/*a*-SiN_x:H superlattice.

The 480-GHz structure can be attributed to a thermal line of the generator spectrum. This line results in a small replica of the 570-GHz structure shifted to lower energies.

In the phonon backscattering spectrum the stop bands give only a weak additional contribution compared with the backscattering signal of the oxygen resonance. This behavior can be explained in the following way. The backscattering of the phonons by the oxygen resonance is a bulk effect, i.e., the backscattering signal corresponds to scattering events in the Si crystal volume close to the generator and the detector junction. The backscattering of the stop-band phonons by the superlattice, however, originates at a much larger distance. Therefore, these backscattered phonons have a high probability for escaping from the crystal into the helium without hitting the detector.

A comparison of the filter characteristics in Figs. 1 and 4 shows that the Si/SiO_2 superlattices, even consisting of a small number of layers, show much sharper stop-band characteristics than *a*-Si:H/*a*-SiN_x:H superlattices. This indicates a possibly higher quality of the evaporated Si/SiO_2 superlattices, mainly with respect to pinholes and the sharpness of the film boundaries.

V. CONCLUSION

We have demonstrated that phonon filters can be prepared very simply with amorphous evaporated Si/SiO_2 multilayer structures as well as with amorphous Si:H/SiN_x:H multilayer structures fabricated by plasma-enhanced chemical vapor deposition. In the case of Si/SiO_2 superlattices excellent stop-band characteristics could be obtained even with a very small period number. The Si:H/SiN_x:H superlattices showed very deep stop bands too. However, the side steepness of the stop bands is reduced. Nevertheless, both amorphous superlattices show significantly "improved" phonon filter properties compared to the MBE-grown GaAs/$Al_xGa_{1-x}As$ filters reported previously.

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