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Raman scattering involving umklapp processes in Si/Ge$_x$Si$_{1-x}$ superlattices

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Raman scattering from folded acoustic phonons in Si/Ge$_x$Si$_{1-x}$ strained-layer superlattices grown by molecular-beam epitaxy is reported. The superlattice periodicity was chosen so that the transferred photon momentum exceeds the minizone momentum π/d, where d is the superperiodicity. The observed peaks agree very well with calculations involving umklapp and folding of the scattered phonons into the minizone.

The formation of a mini-Brillouin zone in a superlattice dramatically changes the phonon spectrum by "folding" the dispersion curves into the minizone. Gaps are created at $q=0$ and at the minizone edge, viz., $q_{\text{mz}}=\pi/d$, where d is the superlattice periodicity. Such folded phonons in GaAs/AlAs superlattices have been studied by acoustic transmission and Raman scattering (RS).[1]

In this Rapid Communication we report a RS study of folded longitudinal-acoustic modes from Si/Ge$_x$Si$_{1-x}$ strained layer superlattices grown by molecular-beam epitaxy (MBE) on (001) Si substrates. This system has less favorable photoelastic constants than in GaAs/AlAs superlattices,[2] but the folded phonons were readily observable (see Fig. 1). We have exploited the possibility of making the minizone wave vector q_{mz} comparable to q_p, the momentum transferred from photon scattering, to investigate umklapp-like RS which is impossible in ordinary crystals. Denoting the minizone-center phonon frequencies by $m\omega_0$, where $m=0,1,2,\ldots$ is the folding index, light scattering peaks should occur at the shifted frequencies $\Delta\omega \simeq m\omega_0 \pm q_p v_{\text{sl}}$, where v_{sl} is the superlattice sound velocity. Thus, each folding index $m \neq 0$ would lead to a doublet, ma and mb corresponding to $-qv_{\text{sl}}$ and qv_{sl}, respectively. The case $m=0$ corresponds simply to Brillouin scattering. Since the observed $\Delta\omega$ were small (<75 cm^{-1}), Rytov's theory[3] of acoustic vibrations in layered media was used to calculate ω_0 and v_{sl}. The observed peaks agree very well with the calculations.

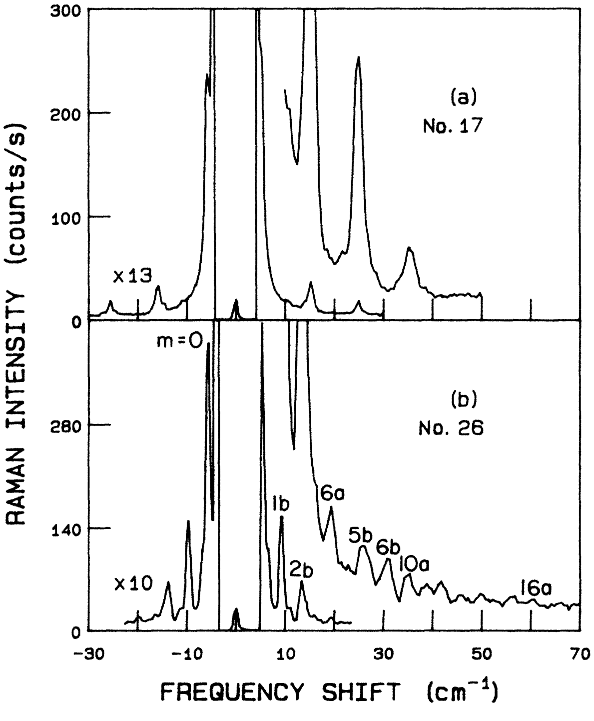

FIG. 1. The unpolarized Raman spectra of Si/Ge$_x$Si$_{1-x}$ superlattices (a) No. 17 and (b) No. 26, recorded at 295 K. See Table I for the labeling of the peaks.

Silicon layers of thickness d_1 were alternated with Ge$_x$Si$_{1-x}$ layers of thickness d_2, with x the Ge composition. A Vacuum Generators V-80 MBE system was used to grow several Si/Ge$_x$Si$_{1-x}$ superlattices of various thicknesses and composition. We discuss here only two samples, viz., Nos. 17 and 26, and will report other aspects of this study elsewhere. In both superlattices the Si/Ge$_x$Si$_{1-x}$ interfaces were commensurate as revealed by cross-sectional transmission electron microscopy (TEM). The epitaxial material was of excellent crystalline quality, as confirmed by Rutherford backscattering. The sharpness of the interfaces are seen from the [110] TEM cross-section micrograph (Fig. 2) of sample No. 26. The accompanying selected area diffraction pattern shows a splitting of the (004) spot along the [001] direction, indicative of tetragonal distortion. Small variation in the layer thickness is seen in Fig. 2. The values of d_1 and d_2 used in the calculation are the mean values for approximately 20 periods of the superlattice sampled by the light (4579 Å). In No. 17, $d_1=225$ Å, $d_2=50$ Å, so that $d=d_1+d_2=275$ Å. The Ge composition $x=0.2$. In No. 26, $d_1=450$ Å, $d_2=200$ Å, and $x=0.45$.

The Raman spectra of the samples placed in a He atmosphere were measured in a 90° scattering geometry with the (001) surface inclined at an angle of 12° to the incident light. The spectra were excited with 300 mW of 4579-Å laser light and analyzed with a Spex 14018 double monochromator at resolutions of 0.8 and 1.6 cm^{-1} for the lower and higher frequencies, respectively. Typical Raman spectra for the two samples are given in Fig. 1. The spectrum of sample No. 17 comprises a number of strong lines accompanied by weaker features, whereas the spectrum of sample No. 26 exhibits a remarkable complexity of detail and in appearance is quite unlike the spectra observed so far for GaAs/AlAs superlattices involving small periodicities.[1]

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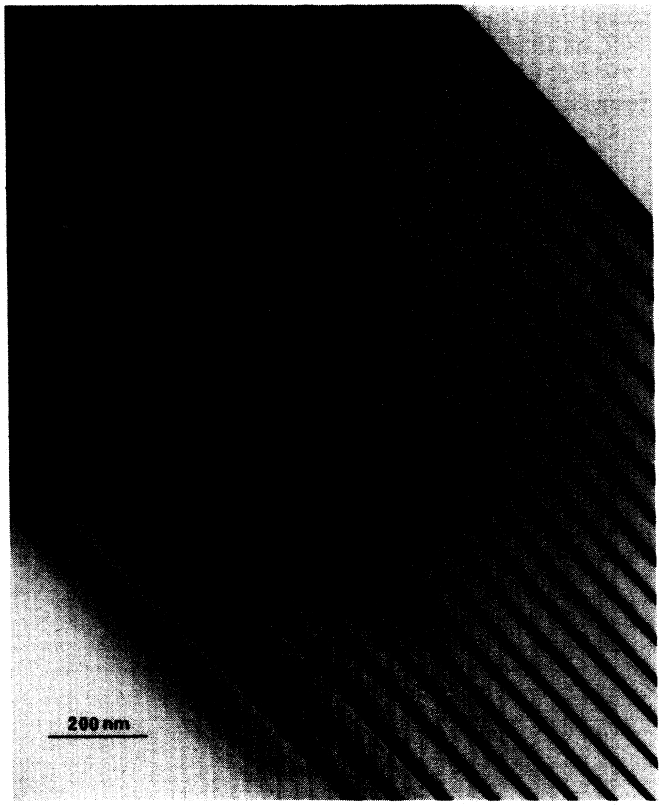

FIG. 2. Cross-sectional TEM micrograph of sample No. 26. Inset: selected area diffraction pattern.

The large refractive index makes the experiment almost backscatteringlike. (The deviations from exact backscattering did not manifest as transverse excitations, although we looked for them.) The photon momentum transfer q_p was deduced from the experimental geometry, the laser wavelength, and the refractive index. Thus for Nos. 17 and 26, $q_p/q_{\mathrm{mz}} \approx 1.09$ and 2.58, respectively, with $q_{\mathrm{mz}} = \pi/d$. In each case light is scattered with $q_p > q_{\mathrm{mz}}$, and hence, the scattering phonon has to be "folded" back (umklapp) into the minizone. This is shown in Fig. 3 for sample No. 17, where the calculated dispersion curves are shown by the solid lines. The experimental points at $q = 0.91$ correspond to the $q_p/q_{\mathrm{mz}} = 1.09$ phonons "folded" back. For example, the doublet corresponding to $m = 3$, i.e., $m\omega_0 \approx 30$ cm^{-1}, is calculated to be at $\Delta\omega = 3\omega_0 \pm q_p v_{\mathrm{sl}}$. This, when folded into the minizone, gives the experimental doublet shown as diamonds at $q/q_{\mathrm{mz}} = 0.91$.

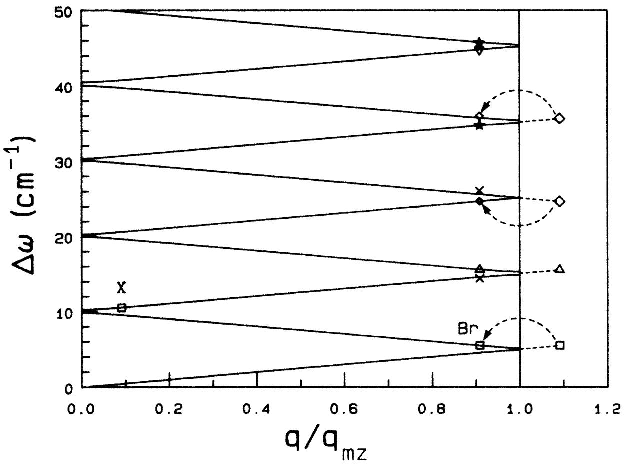

FIG. 3. The calculated dispersion curves and experimental points for sample No. 17.

The observed doublet corresponding to $m = 2$ is shown as two crosses. The very low frequency peak seen at 5.5 cm^{-1} and shown as a square in Fig. 3 and labeled Br is assigned to Brillouin scattering, viz., $m = 0$, with $\Delta\omega = q_p v_{\mathrm{sl}}$. We also observe a weak line at 10.7 cm^{-1}, labeled X in Fig. 3. The exact origin of this line is unclear, but may be assigned to some momentum nonconserving scattering process. We have ascertained that this peak disappears when the scattering momentum q_p is less than q_{mz}, e.g., at a wavelength of 5145 Å, for which $q_p/q_{\mathrm{mz}} = 0.89$. The position of the X peak seems to correspond to $\Delta\omega = (q_p + q_{\mathrm{mz}}) v_{\mathrm{sl}}$ for laser wavelengths where $q_p > q_{\mathrm{mz}}$.

When $q > q_{\mathrm{mz}}$, as in this study, the observed peaks do not form a monotonic progression in the zone-folding index m. This is even more pronounced for example No. 26, for which $q_p/q_{\mathrm{mz}} \simeq 2.58$ for the experimental conditions used. Over thirty RS peaks were observed for sample No. 26 for $\Delta\omega \lesssim 65$ cm^{-1}, and their frequencies agree very well with the calculated spectrum (see Table I). The instrumental error is ± 0.5 cm^{-1}. Table I contains a peak at 16.1 cm^{-1} which does not fit into the zone-folded phonon calculations. However, the observed peak falls (to within experimental error) close to the $m = 4$ zone center (ZC) frequency 16.5 cm^{-1}, and hence, is labeled ZC in the last row of Table I. This peak may arise from some momentum nonconcerving process arising from nonuniformites in the crystal. The intensities are also in qualitative accord with the expected modulation of the form $m^{-2}\sin^2(m\pi d_1/d)$.

TABLE I. Assignment of the first 12 observed Raman scattering peaks for sample No. 26. Peaks with m up to 17 were observed and assigned. Here ma and mb refer to the $\omega_0 m - q v_{\mathrm{sl}}$ and $\omega_0 m + q v_{\mathrm{sl}}$ branches, respectively.

Experiment (cm^{-1})	5.5	6.7	9.4	11.0	13.5	14.4	16.1	17.4	19.6	21.4	· · ·	· · ·	26.3	· · ·	31.2
Calculated (cm^{-1})	5.2	6.9	9.2	10.9	13.2	14.9	· · ·	17.2	18.9	21.2	22.9	25.2	26.9	29.2	30.9
Folding index (m)	0	3*a*	1*b*	4*a*	2*b*	5*a*	ZC	3*b*	6*a*	4*b*	7*a*	5*b*	8*a*	6*b*	9*a*

In summary, the unique physical properties of thick-layer superlattice structures have enabled extraordinary measurements of the Raman spectrum of a solid at wave vectors *outside* its first Brillouin zone. The corresponding positions within this zone are found by folding a given wave vector $q/q_{\mathrm{mz}}=n+f$, where n is an integer and f the fractional part, to give $\tilde{q}/q_{\mathrm{mz}}=R+(-1)^n f$, where R is 0 or 1 for even or odd n, respectively. This is the first time such umklapp processes have been observed in a light scattering experiment.[4]

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[1] C. Colvard, T. A. Gant, M. V. Klein, R. Merlin, R. Fischer, H. Morkoc, and A. C. Gossard, Phys. Rev. B **31**, 2080 (1985), and references therein.

[2] S.-Y. Ren and W. A. Harrison, Phys. Rev. B **23**, 762 (1981), see their Table III.

[3] S. M. Rytov, Akust. Zh. **2**, 71 (1956) [Sov. Phys. Acoust. **2**, 68 (1956)].

[4] See R. Merlin, C. Colvard, M. V. Klein, H. Morkoc, A. Y. Cho, and A. C. Gossard, Appl. Phys. Lett. **36**, 43 (1980); regarding the earlier work of G. A. Sai-Halasz, A. Pinczuk, P. Y. Yu, and L. Esaki, Solid State Commun. **25**, 381 (1978).

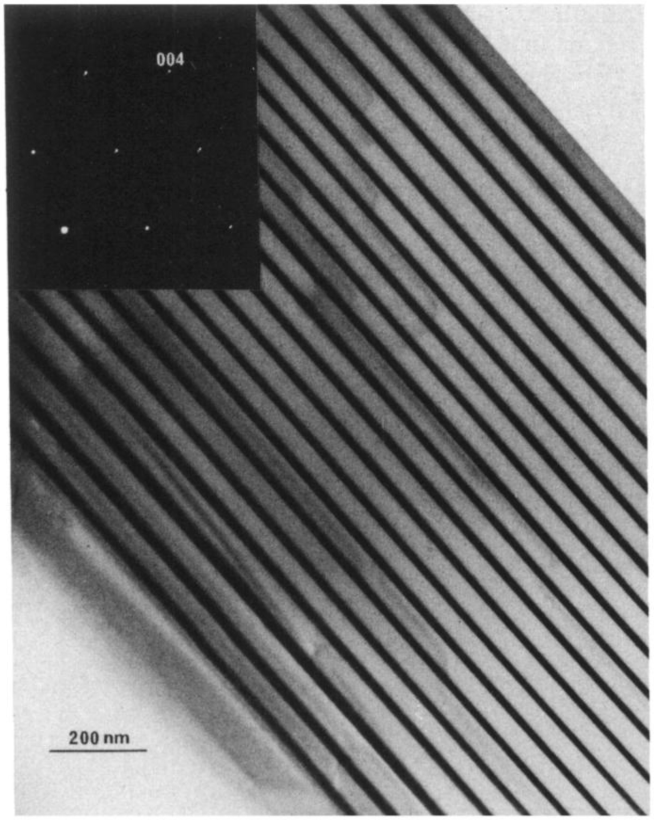

FIG. 2. Cross-sectional TEM micrograph of sample No. 26. Inset: selected area diffraction pattern.