Raman scattering from TO phonons in $(GaAs)_n/(AlAs)_n$ superlattices

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$(GaAS)_n/(AlAs)_n$ superlattices with $n=4$, 6, and 8 grown by molecular-beam epitaxy on (001)-oriented GaAs substrates were investigated by Raman scattering. In a strict backscattering geometry, confined TO-phonon modes with E symmetry are Raman forbidden. However, the effects due to near-Brewster-angle incidence and a large aperture of the scattering-light collecting lens create a small wave-vector component along the (110) orientation, and thus induce a Raman activity of TO phonons. When we take $X\|[1\bar{1}0]$, $Y\|[110]$, and $Z\|[001]$, in the near-$Z(YX)\bar{Z}$ backscattering configuration confined LO-phonon modes are Raman inactive. Using this configuration, we have for the first time observed both GaAs-like and AlAs-like confined TO-phonon modes at room temperature and under off-resonance conditions.

Since Esaki and Tsu[1] proposed artificial superlattice structures, considerable efforts have been devoted to the investigations of the properties of semiconductor superlattices. The early studies were mainly concerned with the electronic properties with particular emphasis on the fundamental physics and device applications. In recent years, the vibrational properties of superlattices have also attracted much interest. Most of the studies in this respect were concentrated on Raman scattering studies of folded LA-phonon and confined LO-phonon modes (see review papers, Refs. 2 and 3). Also Raman-scattering measurements of folded TA-phonon modes were reported.[4,5] As to the Raman-scattering studies of confined TO phonons, to our knowledge there has only been the report by Sood *et al.*[6] on GaAs-like confined TO-phonon modes observed at 10 K and under extreme resonance conditions, but no AlAs-like confined TO-phonon modes have been reported yet. Raman scattering measurements of TO phonons are difficult because in backscattering geometry the TO-phonon modes with E symmetry are Raman forbidden for the GaAs/AlAs superlattices grown along the (001) orientation. On the other hand, the TO phonons are less dispersive than LO being generally within a narrow range of about 20 cm^{-1}, and thus difficult to observe by Raman scattering.[7,8]

In this paper, we report the first observation of both GaAs-like and AlAs-like confined TO-phonon modes in GaAs/AlAs superlattices at room temperature and under off-resonance conditions. The three $(GaAs)_n/(AlAs)_n$ superlattice samples used in this experiment were grown by alternatively depositing the constituent GaAs and AlAs layers on (001)-oriented GaAs substrates by molecular-beam epitaxy.[9] The constituent-layer thickness is represented by n, the number of monolayers. Each monolayer is given by $a_0/2=2.83$ Å for GaAs and AlAs (a_0 is the lattice constant). In the three samples used in our measurements, n is equal to 4, 6, and 8, respectively, as determined by double-crystal x-ray diffraction.[9,10] The Raman spectra were recorded with a Jobin-Yvon T800 triple-grating laser Raman spectrometer at room temperature. To distinguish the confined TO-phonon modes distributed in a narrow frequency range, a sufficient spectral resolution is necessary. The 5145-Å line of a Spectra-Physics Model 165 Ar-ion laser was focused on the sample surfaces with a typical power of less than 100 mW. The temperature increase of the samples resulting from the exciting light was about 40 K as estimated from the intensity ratio of the Stokes and antiStokes LO_1 modes. This temperature increase can result in a frequency decrease of about 0.6 cm^{-1}, as expected from the temperature dependence of optical phonons in bulk GaAs. The real temperature increase and therefore the frequency decrease might, of course, be slightly different since the details of the structures and the quality of superlattices are different from those of bulk GaAs.

In $(GaAs)_n/(AlAs)_n$ superlattices with D_{2d} point-group symmetry, there are n doubly degenerated confined TO-phonon modes as denoted by TO_m ($m=1,2,\ldots,n$). In a strict backscattering geometry, both incident- and scattered-light wave vectors are along the (001) orientation, and TO-phonon modes are thus Raman inactive. However, the effects due to the near-Brewster-angle incidence and a large aperture of the scattering-light-collecting lens create a small component of the wave vector perpendicular to the (001) orientation, which can thus induce the Raman activity of the TO modes. In this configuration, because of the large refractive index in GaAs and AlAs layers, the refractive angle within the samples is only about 10°, and it can therefore still be considered as a near-backscattering geometry.

In Fig. 1 we show the Raman spectra of the LO- and TO-phonon modes confined in the GaAs layers of the $(GaAs)_4/(AlAs)_4$ superlattice sample. The LO_m- and TO_m-phonon modes were observed with the near-$Z(YY)\bar{Z}$ and near-$Z(YX)\bar{Z}$ backscattering configurations, respectively, where $X\|[1\bar{1}0]$, $Y\|[110]$, and

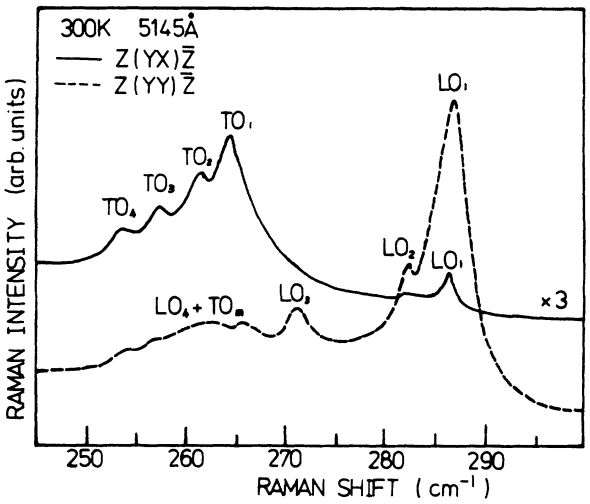

FIG. 1. Raman spectra of GaAs-like LO_m- and TO_m-phonon modes in a $(GaAs)_4/(AlAs)_4$ superlattice sample with near-$Z(YY)\bar{Z}$ and near-$Z(YX)\bar{Z}$ back scattering configurations, where $X\|[1\bar{1}0]$, $Y\|[110]$, and $Z\|[001]$.

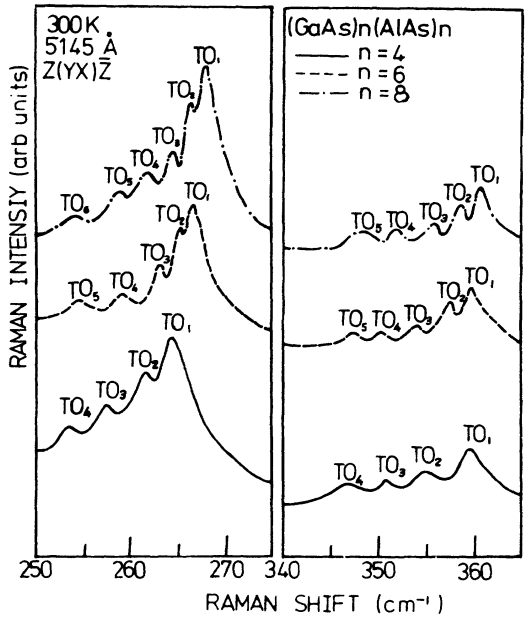

FIG. 2. Raman spectra of GaAs-like and AlAs-like TO_m-phonon modes in $(GaAs)_n/(AlAs)_n$ superlattice samples with $n=4$, 6, and 8, near the $Z(YX)\bar{Z}$ backscattering configuration, where $X\|[1\bar{1}0]$, $Y\|[110]$, and $Z\|[001]$.

$Z\|[001]$. As mentioned in a previous paper,[11] for the $Z(YY)\bar{Z}$ configuration both even and odd LO_m-phonon modes are Raman active. As indicated by the dashed line of Fig. 1, the LO_1-, LO_2- and LO_3-phonon modes can be identified unambiguously, while the LO_4 mode appears only as a broad and rather weak band because of the overlap with the TO_m-phonon modes in the range 250–270 cm^{-1}. In this configuration, the intensities of TO_m-phonon modes are so weak that the TO_1 mode, the strongest one among all TO_m modes, is shown only with nearly the same intensity as that of LO_4, the weakest one of the LO_m modes, and they are thus difficult to be resolved from each other. This problem becomes more difficult in the case of samples with thicker constituent layers. For instance, the phonon modes LO_5 (269 cm^{-1}) and LO_6 (262 cm^{-1}) in the sample of $n=6$, or LO_6 (271.5 cm^{-1}), LO_7 (266 cm^{-1}), and LO_8 (261.5 cm^{-1}) in the sample of $n=8$, are just in the same range as the TO_m-phonon modes. They are therefore ambiguous to be distinguished from each other. As a matter of fact, the GaAs-like TO_m modes observed by Sood *et al.*[6] at 10 K and under extreme resonance conditions were just superimposed on the lower-energy tail of the stronger LO_6 mode.

In order to avoid the interference from LO_m-phonon modes, a near-$Z(YX)\bar{Z}$ backscattering configuration was thus employed. Since all LO_m-phonon modes are Raman forbidden in this configuration,[11] as shown in Fig. 1 by the solid line, all TO_m-phonon modes are observed quite clearly, while all LO_m-phonon modes, except for a weak leakage of the LO_1 mode, are suppressed. In Fig. 2, the Raman spectra of both the GaAs-like and AlAs-like TO_m-phonon modes of $(GaAs)_n/(AlAs)_n$ superlattices with $n=4$, 6, and 8 are depicted. All four GaAs-like and AlAs-like TO_m modes are detected unambiguously from the sample with $n=4$. However, a few higher-order

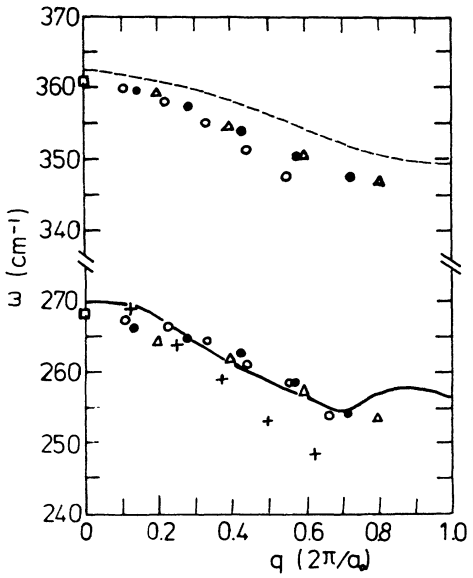

FIG. 3. Measured GaAs-like and AlAs-like TO_m-phonon mode frequencies in $(GaAs)_n/(AlAs)_n$ superlattice samples vs $q=[m/(n+1)](2\pi/a_0)$. △, $n=4$; ●, $n=6$; ○, $n=8$, Raman-scattering measurement data (present work). +, $n=7$, Raman-scattering measurement data by Sood *et al.* (Ref. 6). □, TO(Γ) frequencies of bulk GaAs and AlAs (Ref. 15) from Raman-scattering measurements. ——, phonon-dispersion curve of bulk GaAs (Ref. 14) from neutron-scattering measurement data. — — —, calculated phonon-dispersion curve of bulk AlAs.

TO_m-phonon modes are not observed from the samples with $n=6$ and 8, partly due to the low signal-to-noise ratios and partly due to superpositions between higher-order TO_m-phonon modes resulting from the flattening of the dispersion curves at large q values.[7,8]

Based on the slab model, Sood *et al.*[6] suggest the correspondence between the confined-mode and slab-mode frequencies. The authors plotted the confined LO_m-mode frequencies versus $q=m\pi/d_1=(m/n)(2\pi/a_0)$, and they found a good agreement with the phonon-dispersion curve of GaAs bulk material, except for the range of large q values where considerable discrepancies appeared. Subsequently, Jusserand and Paquet[12] pointed out that the optical-phonon modes in superlattice structures are confined modes rather than slab modes. Based on a linear-chain model and using a vanishing penetration depth into the AlAs layers, they showed that the frequencies of optical phonons confined in GaAs layers correspond to those of optical phonons in GaAs bulk material at $q=[m/(n+1)](2\pi/a_0)$ instead of $q=(m/n)(2\pi/a_0)$. $(n+1)$ may be replaced by n only when n is large enough.[13] In Fig. 3, the frequencies of TO_m-phonon modes versus $q=[m/(n+1)](2\pi/a_0)$ together with the phonon-dispersion curves of bulk GaAs and AlAs are plotted. For bulk GaAs we use the most recent and very accurate data obtained by neutron scattering[14] instead of those obtained previously at room temperature.[7] For bulk AlAs, on the other hand, neutron-scattering data are not yet available, and thus we use the theoretical phonon-dispersion curve calculated by Barker *et al.*[8] Our present experimental data obtained from $(GaAs)_n/(AlAs)_n$ superlattices are in good agreement with those of the constituent bulk materials. In addition, as seen in Fig. 3, in the range of $q>0.6(2\pi/a_0)$ the distributions of the phonon-dispersion curves of bulk materials and our experimental data are flattened and become nearly nondispersive. As a matter of fact, a few higher-order TO_m modes in the samples with $n=6$ and 8 cannot be distinguished because the frequencies of these modes are located very close to each other.

In conclusion, our investigations have shown that the Raman-scattering measurements of superlattice structures provide an effective method to determine the phonon-dispersion relations of the constituent bulk materials. This is particularly important for some materials which are not stable in air, such as AlAs, or which can be prepared only in the form of thin films because of the limitation of crystal-growth techniques, such as CdS. For these materials neutron-scattering data are not available. On the other hand, the discrepancies between the calculated phonon-dispersion relations for these materials are quite large, especially in the near-Brillouin-zone-edge region. For instance, the calculated frequency of $TO(X)$ in AlAs is either larger than that of $TO(\Gamma)$ (Ref. 16) or lower than 300 cm^{-1}.[17] The experimental results obtained from Raman-scattering measurements have thus become particularly meaningful.

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