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Brief Reports

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Confined optical phonons in a GaAs single quantum well in a GaAs/$Al_xGa_{1-x}As$ heterostructure

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We report the observation of confined longitudinal-optical (LO) phonons in the first-order Raman spectrum of a GaAs/$Al_xGa_{1-x}As$ single quantum well. They are observed when the energy of the scattered radiation comes into the resonance with the electronic transitions of the quantum well, the resonance being tuned with temperature or with a tunable dye laser. The confined LO phonons observed in the first-order Raman spectrum have wave vectors extending to a significant fraction of the Brillouin zone and their frequencies agree well with those deduced from the bulk dispersion curve.

There is considerable current interest in the vibrational properties of semiconductor heterostructures.[1-3] In the context of the vibrational dispersion relations of the constituent components of semiconductor heterostructures, many fundamental questions arise, e.g., the propagation or the confinement of the vibrational modes depending on the matching of the dispersion curves of the constituents. In multiple-quantum-well (MQW) structures, it has been observed[2,3] that acoustic phonons, whose dispersion curves in the two constituents overlap over a wide range of frequencies, propagate through both layers exhibiting zone-folding effects due to the new periodicity. On the other hand, optical phonons show "confinement"[2-4] when the dispersion curves do not overlap and "propagation" when they do. In contrast to acoustic phonons, zone-folding effects are not observed for propagating optical phonons because of the highly dispersive character of the optical modes.[3]

It appeared to us particularly attractive to investigate the vibrational modes in a single-quantum-well (SQW) structure to complement the studies on MQW structures.[2-4] In effect the problem reduces to that of a single thin slab. In the present Brief Report we report the observation of confined LO phonons even in GaAs/$Al_xGa_{1-x}As$ SQW's provided resonance conditions are appropriately exploited.

Single quantum wells of GaAs/$Al_xGa_{1-x}As$ heterostructures have been grown by molecular-beam epitaxy (MBE), the layer being perpendicular to [001]. SQW I consists of a 46-Å-thick GaAs well with $Al_xGa_{1-x}As$ ($x=0.3$) barrier layers on either side of the well, whereas in SQW II a 31-Å GaAs well is sandwiched between $Al_xGa_{1-x}As$ ($x=0.28$) barrier layers. Raman spectra were obtained in the backscattering configuration using the 6764- or 7525-Å lines of a Kr^+ laser and the various wavelengths provided by a tunable dye laser operating with pyridin 1. Low-temperature measurements were carried out in either a variable-temperature stainless-steel cryostat or an optical magnet cryostat equipped with a superconducting solenoid providing magnetic fields up to 60 kG. Scattered light was spectrally analyzed with a computer-controlled double monochromator and detected with a standard photon-counting unit.[3]

It is useful to recall that the confined modes of a slab consisting of N monolayers of total thickness $Na/2$ are actually the N discrete modes with frequencies corresponding to N discrete points with their wave vectors, $2n\pi/Na$, given by the bulk dispersion curves and spanning the Brillouin zone.[4] Here a is the lattice constant and $n=1,2,\ldots,N$. However, if the GaAs slab has AlAs or $Al_xGa_{1-x}As$ slabs on either side, as in the case of our SQW's, it has been argued[5] that the atomic displacements do not vanish at the boundary of the GaAs slab, but vanish at the first Al atoms in the neighboring slabs, thus making the effective slab thickness $N+1$ monolayers in the context of phonon confinement.

Figure 1 shows the Raman spectrum of SQW I at 90

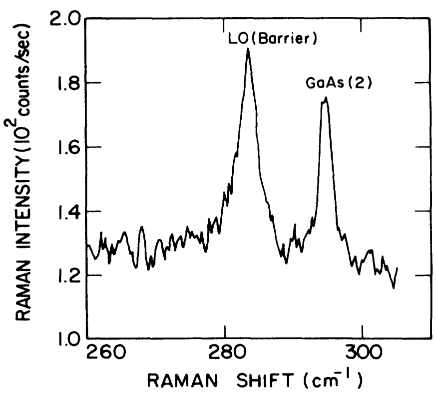

FIG. 1. Raman spectrum of the GaAs SQW at $T=80$ K in the $z(yy)\bar{z}$ configuration using the 6764-Å line of a Kr^+ laser. Peak labeled LO(barrier) is the GaAs-like LO phonon of the $Al_xGa_{1-x}As$ barrier. The peak labeled GaAs(2) is the $n=2$ confined LO phonon of the GaAs slab.

K using the 6764-Å line of the Kr^+ laser. Two peaks are observed in the GaAs-like phonon region. The peaks have a polarization identical to that of the incident photon. The peak labeled GaAs(2) is the GaAs LO phonon originating in the SQW; its polarization suggests that it belongs to the A_1 symmetry of the D_{2d} point-group symmetry of the SQW. Hence it should be assigned to the $n=2$ confined mode of the 46-Å GaAs layer.[4] The signal is very weak since only a single layer is involved. The peak labeled LO(barrier) is the GaAs-like LO phonon arising from the $Al_xGa_{1-x}As$ barrier. Its frequency matches well with the GaAs-like phonon of bulk $Al_xGa_{1-x}As$.[6]

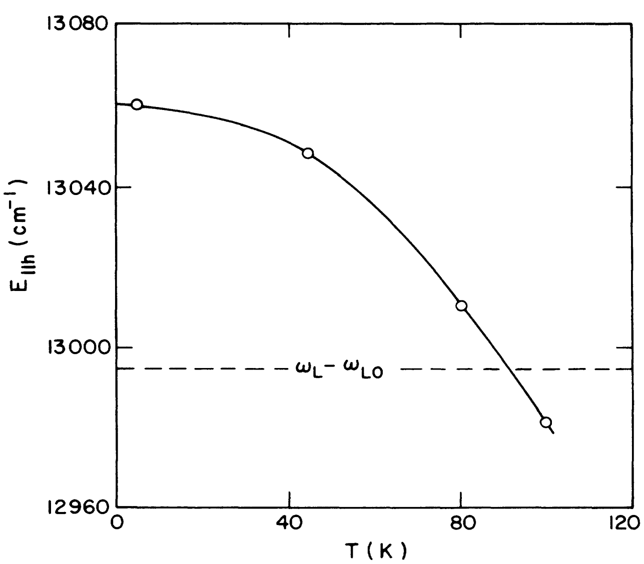

FIG. 2. Temperature dependence of the E_{11h} luminescence energy. Horizontal dashed line labeled $\omega_L - \omega_{LO}$ is the energy at which out resonance will occur for the 7525-Å line of the Kr^+ laser with the LO phonon of GaAs.

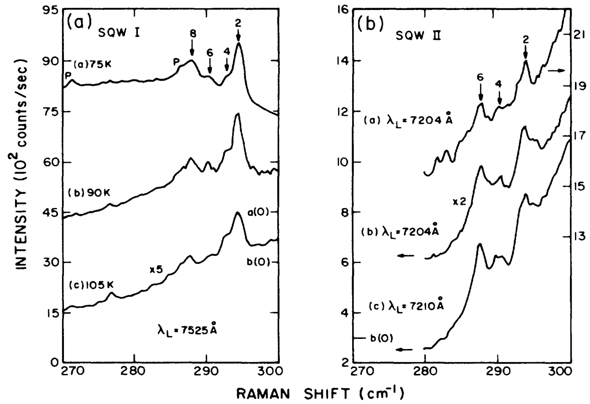

FIG. 3. (a) Raman spectra of the GaAs SQW I at different temperatures in the $z(yy)\bar{z}$ configuration using the 7525-Å line of the Kr^+ laser. (a) $T=75$ K, (b) $T=90$ K, and (c) $T=105$ K. The four Raman peaks labeled 2, 4, 6, and 8 are the confined LO phonons of the GaAs slab. The peaks labeled P are plasma lines. (b) The confined LO phonons in SQW II. (a) $T=10$ K, $H=0$, $\lambda_L=7204$ Å, (b) $T=5$ K, $H=60$ kG, $\lambda_L=7204$ Å, and (c) $T=5$ K, $H=60$ kG, $\lambda_L=7210$ Å; in (b) and (c) the angle between the magnetic field and [001] is 45°.

Since the Raman signal arises from a single 46-Å layer of GaAs, one can hope to see the higher-order confined phonons only under strong resonant conditions. The conduction subband 1 to heavy-hole subband $1h$ luminescence energy[7] of the SQW, E_{11h}, can be varied by changing the temperature. Figure 2 shows the variation of E_{11h} as a function of temperature. Note that one

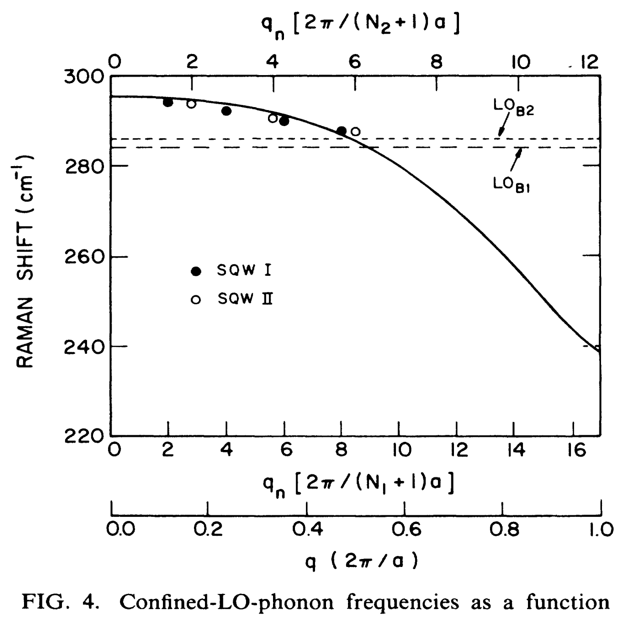

FIG. 4. Confined-LO-phonon frequencies as a function of the quantized wave vector $q_n=2n\pi/(N+1)a$ in SQW I and SQW II. The frequencies of SQW I are those for $T=75$ K. The continuous curve is the bulk dispersion curve of the GaAs LO phonon in the [001] direction calculated in Ref. 10, adjusted for $T=5$ K. The dashed lines are the GaAs-like zone-center LO frequencies of the $Al_xGa_{1-x}As$ barrier: LO_{B1} is for SQW I and LO_{B2} is for SQW II.

can tune E_{11h} across the energy of the photon scattered by the GaAs LO phonon using the 7525-Å line of the Kr^+ laser, i.e., by "temperature tuning" the resonance.[8] Note also that strong resonance is expected between 80 and 100 K.

Figure 3(a) shows the Raman spectra of SQW I at different temperatures using the 7525-Å line of the Kr^+ laser. The Raman peaks now ride over the E_{11h} luminescence fulfilling the condition of "out resonance," i.e., the energy of the scattered light equals an electronic transition energy. Four Raman peaks labeled 2, 4, 6, and 8 are seen between the GaAs LO and the GaAs-like LO frequencies and are assigned to LO phonons confined to the GaAs layer. All the peaks have a polarization identical to that of the incident photon indicating that these peaks have A_1 symmetry and should correspond to the confined modes of the GaAs layer with even n values,[4] $n=2$, 4, 6, and 8. Under the resonance condition, only modes with even n values dominate the Raman spectrum.

Similarly in SQW II, the confined LO phonons with $n=2$, 4, and 6 are observed under out-resonance condition now obtained by selectively tuned incident photon energy, as shown in Fig. 3(b). Although the luminescence peak does not shift with the applied magnetic field, we noticed that the intensity of phonons are resonantly enhanced more efficiently under the magnetic field.[9]

Finally, in Fig. 4 we show the observed confined-LO-phonon frequencies as a function of quantized wave vector $2n\pi/(N_{1,2}+1)a$ along with the calculated bulk dispersion curve[10] for GaAs at 5 K. One can see that there is an excellent agreement between the frequencies of the confined LO phonon and those expected from the bulk dispersion curve. It is important to point out that in a $GaAs/Al_xGa_{1-x}As$ SQW the LO phonon does not remain confined below the GaAs-like LO frequency of the $Al_xGa_{1-x}As$ barrier which is shown as a dashed horizontal line in Fig. 4.

The present study shows that, with a set of suitably designed SQW's, and appropriately exploiting the resonance Raman effect associated with the quantum well, one can observe the first-order Raman line associated with the phonons over a large portion of the dispersion curve; this is in contrast to the first-order Raman scattering in the bulk, where only optical phonons with wave vectors close to the zone center are observed. The higher-frequency precision as well as applicability even to Cd-based constituents make Raman scattering from appropriately fabricated heterostructures an attractive alternative to inelastic neutron scattering for determining dispersion curves.

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