



## Raman scattering from confined phonons in GaAs/AlGaAs quantum wires

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We report on photoluminescence and Raman scattering performed at low temperature ( $T = 10$  K) on GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As quantum-well wires with effective wire widths of  $L = 100.0$  and  $10.9$  nm prepared by molecular beam epitaxial growth followed by holographic patterning, reactive ion etching, and anodic thinning. We find evidence for the existence of longitudinal optical phonon modes confined to the GaAs quantum wire. The observed frequency at  $\omega_{L10} = 285.6$  cm<sup>-1</sup> for  $L = 11.0$  nm is in good agreement with that calculated on the basis of the dispersive dielectric continuum theory of Enderlein<sup>†</sup> as applied to the GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As system. Our results indicate the high crystalline quality of the quantum-well wires fabricated using these techniques.

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**Key words:** confined phonons, quantum wires.

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### 1. Introduction

With the recent improvements in microfabrication technology of nanostructures the growth of semiconductor systems with reduced dimensionality has been actively pursued in recent years. The ability to realize such quantum wires can lead to new flexibility in tailoring optical and transport properties with respect to novel physical phenomena as well as potential applications. Due to significant modifications in the density of states, improved laser and high-speed device performance using quantum-well wires (QWW) as the active region has been predicted [1–5]. Among the various systems under current investigation, heteroepitaxial GaAs/AlGaAs quantum wires have attained considerable theoretical and experimental attention. The presence of heterointerfaces with large changes in the dielectric constant can produce confinement of the optical phonon modes as well as localization in the vicinity of interfaces known as the interface and surface optical modes [5]. Experimental evidence for surface phonons in cylindrical GaAs quantum wires was found by Watt *et al.* [6].

Several macroscopic theoretical models have been proposed to deal with the allowed phonon modes in quantum wire structures. In the dielectric continuum model [8] confined and surface (interface) modes are

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determined by the electrostatic boundary conditions. The hydrodynamic continuum model [9, 10] makes use of the mechanical boundary conditions to obtain the guided modes. A semimicroscopic model developed by Huang and Zhu [10] also addresses the mechanical boundary conditions. However, in spite of these theoretical efforts, there has been no experimental verification up to now due to many difficulties fabricating high-quality quantum wires.

In this paper, we present results from the realization of GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As QWW arrays with effective wire widths of  $L = 100.0$  and  $10.9$  nm, and report their low-temperature (10 K) photoluminescence and Raman study. The photoluminescence spectra show blue energy shifts while in the Raman spectra a new phonon mode appears. We will argue that this new peak could possibly be identified by using the dispersive dielectric continuum model developed recently [7].

## 2. Results and discussion

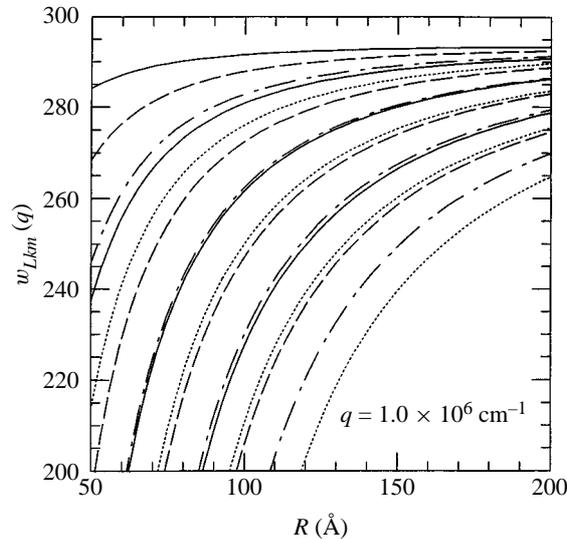
The dielectric continuum model for circular quantum wires as developed by Enderlein [7] makes use of the generalized Born–Huang equation with a hermiticity condition for the dynamical operator. The dispersion relation for the zone-center longitudinal optical [LO( $\Gamma$ )] phonon modes confined to the GaAs/AlGaAs quantum wire in the frame of this model is given by

$$\omega_{Lkm}^2(q) = \omega_L^2 - \beta_L^2 \left[ q^2 + \left( \frac{x_{km}}{R} \right)^2 \right], \quad (1)$$

where  $\omega_L$  is the bulk LO( $\Gamma$ ) phonon frequency,  $\beta_L$  is the velocity parameter,  $x_{km}$  denotes the  $k$ th root of the Bessel function  $J_m(x)$ , and  $R$  is the radius of the quantum wire. In Fig. 1 we depict the frequencies of the longitudinal optical (LO) phonon modes confined to GaAs in GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As quantum wire as a function of wire radius  $R$ , calculated for wavevector  $q = 1.0 \times 10^6 \text{ cm}^{-1}$ . For the bulk GaAs LO( $\Gamma$ ) phonon frequency  $\omega_L$ , we take the value  $294.5 \text{ cm}^{-1}$ , observed in this experiment at  $T = 10$  K. The velocity parameter for GaAs is  $\beta_L = 4.73 \times 10^3 \text{ m s}^{-1}$ .

Our QWWs were fabricated starting from a 20 nm period GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As superlattice grown by molecular beam epitaxy on a [001] direction oriented semi-insulating GaAs substrate. Each structure consisted of 30 periods of 10 nm GaAs layers, separated by 10 nm Al<sub>0.3</sub>Ga<sub>0.7</sub>As layers lightly doped with Si ( $n = 2.5 \times 10^{11} \text{ cm}^{-2}$  was found from Hall measurements at 2 K). A mask was prepared (for subsequent etching) consisting of photoresist lines with 200 nm periodicity and oriented parallel to  $[\bar{1}10]$  direction. The wires were prepared by high resolution laser interference lithography and reactive ion etching and further thinned by anodic oxidation techniques. On each QWW sample a small unpatterned part was left as reference. The formation of the wire structures were checked by using a scanning electron microscope. The electron micrograph cross sections of the cleaved edge of the GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As QWW array demonstrated the formation of wires with the lateral width of  $L = 100.0$  for thin, and  $\sim 11.0$  to  $13.0$  nm for ultrathin wires, respectively. The samples were mounted on the cold finger of a closed cycle cryostat and kept at  $T = 10$  K. The photoluminescence and Raman spectra were obtained in the backscattering  $z(x\bar{x})\bar{z}$  geometry, where  $z$  and  $x$  are along the [001] and [100] directions. The 457.9 and 514.5 nm emission lines of an Ar<sup>+</sup>-ion laser were used as the excitation source.

The sharp line at 1.552 eV detected in the photoluminescence spectra from the QWW sample from the 100 nm width reflects the confinement due to quantum wells. The full width at half maximum (FWHM) of the photoluminescence band is about 38 meV. This is an acceptable value, taking into account the lightly doped nature of the quantum wires. More importantly, this line only broadens to 39.5 meV in the thinnest wire. These data indicate that the fluctuations in wire size are small demonstrating the good quality of the wire fabrication process. Furthermore, as we thin down the wire width, we obtain an approximately  $\times 1.5$  improvement in the recombination efficiency for the ultrathin sample despite the small active QWW area which occupies less than 1% of the patterned area. We also observe a shift in the peak position by 8.4 meV for the ultrathin QWW sample, indicating the effect of confinement due to quantum wire information.

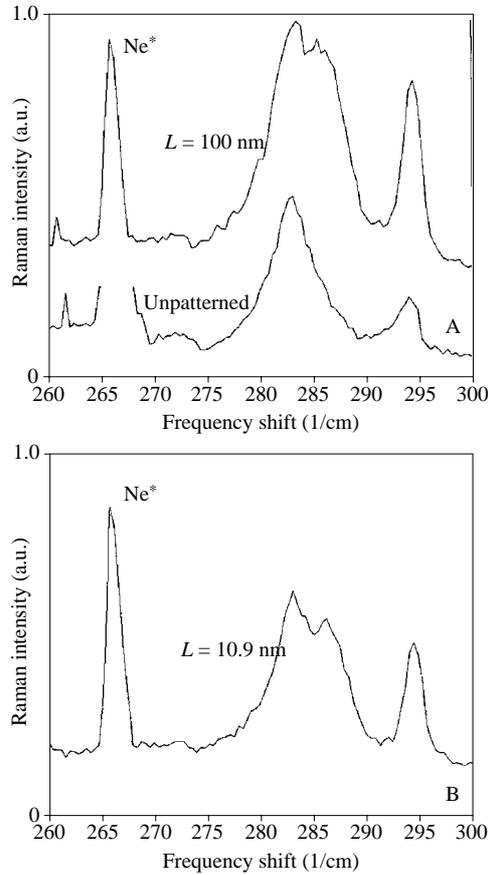


**Fig. 1.** Dispersion of the LO phonon modes confined to GaAs in GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As quantum wires calculated for wave vector  $q = 1.0 \times 10^6 \text{ cm}^{-1}$  by using eqn (1). The modes corresponding to  $m = 0, 1, 2,$  and  $3,$  are shown by solid, dashed, dash-dotted, and dotted lines, respectively. For a given mode  $m,$  from top to bottom,  $k = 1, 2,$  and  $3,$  respectively.

Figure 2 shows the Raman spectra of the GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As QW samples; (A) is unpatterned and  $L = 100.0 \text{ nm},$  (B) is patterned with wire width  $L = 10.9 \text{ nm}$  in the range of TO and LO phonons of GaAs. In this geometry, zinc-blende structures oriented perfectly parallel to [001] direction, scattering by zone-center LO phonons is dipole forbidden. On the other hand, violations of the selection rule may occur due to lowering of the symmetry and lattice disorder such as surface roughness, dislocations, and inhomogeneous strain and impurities. Thus TO phonons may become active in the Raman scattering indicating crystalline disorder. In the spectra of the unpatterned and  $L = 100.0 \text{ nm}$  QWW samples (spectra A), the Stokes component arises from LO phonons of GaAs at  $294.5 \text{ cm}^{-1},$  and GaAs-like LO phonons of Al<sub>0.3</sub>Ga<sub>0.7</sub>As at  $283.7 \text{ cm}^{-1}.$  We note that in our long-period superlattices the phonon quantization energies are so small that higher order confined LO phonons are not observed in the scattering spectra taken with spectral resolution of  $1.4 \text{ cm}^{-1}.$  It is very important that the positions of the LO phonon line of Al<sub>0.3</sub>Ga<sub>0.7</sub>As at  $283.7 \text{ cm}^{-1}$  and LO phonon line of GaAs at  $294.5 \text{ cm}^{-1}$  do not exhibit any shifts within  $\pm 0.05 \text{ cm}^{-1}$  in the spectra if unpatterned samples are used. Scattering by TO phonons at  $\sim 272 \text{ cm}^{-1}$  does not appear in any of the spectra collected.

The most striking feature of the Raman scattering spectra of our GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As QWW is the observation of a new line at  $285.6 \text{ cm}^{-1}$  which appears only in the spectra of the ultrathin QWW sample. This frequency is very close to the calculated values of the frequencies of LO phonon modes confined to GaAs for the lowest order mode  $m = 0$  and  $k = 1$  (topmost curve in Fig. 1). Using the value of  $\omega_{L10} = 285.6 \text{ cm}^{-1}$  we can also obtain directly from eqn (1) the effective wire width of  $L = 10.9 \text{ nm}.$  This value is in a reasonable agreement with our estimation from the electron micrograph cross-section data. In this connection, it is interesting to note that this line appears in all spectra of our QWW samples, taken at several points with  $\sim 250 \mu\text{m}$  step within a  $1.5 \times 1.5 \text{ (mm)}^2$  area showing the same frequency (within  $\pm 0.05 \text{ cm}^{-1}$ ) which also reflects the high homogeneity of the fabricated QWWs.

In summary, we realized GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As QWWs fabricated by high resolution laser lithography followed by reactive ion etching and by anodic oxidation of  $20 \text{ nm}$  period superlattices grown by MBE. The new peak observed at  $\omega_{L10} = 285.6 \text{ cm}^{-1}$  in the Raman spectra of QWW with  $L = 10.9 \text{ nm}$  is consistent with the calculated value of the LO phonon mode confined to GaAs in GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As cylindrical QW as described



**Fig. 2.** Raman spectra of GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As QWW samples: A, unpatterned and wire width  $L = 100.0$  nm; and B, patterned with wire width  $L = 10.9$  nm.

by the dispersive dielectric continuum model for the lowest mode. This good agreement as well as the absence of scattering by TO( $\Gamma$ ) phonons in the spectra of all QWW samples indicate their high crystalline quality.

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