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Optical phonon modes in InGaN/GaN dot-in-a-wire heterostructures grown by molecular beam epitaxy

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We report on the studies of optical phonon modes in nearly defect-free GaN nanowires embedded with intrinsic InGaN quantum dots by using oblique angle transmission infrared spectroscopy. These phonon modes are dependent on the nanowire fill-factor, doping densities of the nanowires, and the presence of InGaN dots. These factors can be applied for potential phonon based photodetectors whose spectral responses can be tailored by varying a combination of these three parameters. The optical anisotropy along the growth (*c*-) axis of the GaN nanowire contributes to the polarization agility of such potential photodetectors. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4798245]

InGaN/GaN dot-in-a-wire nanostructures have received increasing attention in the past few years due to the pressing need for visible light emitting diodes with high internal quantum efficiency.[1,2] Improved technologies in the growth and fabrication processes and the ability to tune the optical properties by implementing ternary alloy structures have given impetus to this foray. High performance bias selectable photodetectors based on quantum dots embedded in quantum well (dot-in-a-well) structures have been demonstrated previously.[3,4] In this article, we discuss the infrared optical properties of these dot-in-a-wire nanostructures to determine their candidacy as infrared photodetectors.

Vertically aligned InGaN/GaN dot-in-a-wire heterostructures were grown on a semi-insulating Si(111) substrate by radio frequency plasma assisted molecular beam epitaxy (Fig. 1). GaN nanowires of 200 nm are first grown at ~750 °C. Nitrogen flow rate of 1 sccm and forward plasma power of ~400 W were kept stable during the nanowire growth.

The fill factor of the nanowires increases radially from the center to the edge of the wafer as seen in Figure 2. This is achieved by introducing a temperature gradient radially decreasing from ~750 °C at the wafer center to ~700 °C at the edge during the nanowire growth process. The wurtzite structured nanowires have their *c*-axis parallel to the growth direction and perpendicular to the substrate surface. Using a growth technique described elsewhere,[2] ten self-assembled intrinsic InGaN quantum dots are then grown at ~600 °C and capped by ~5–10 nm GaN layer doped with Mg after each InGaN dot. Approximately, 20% indium fraction is attained by appropriately controlling the In and Ga beam fluxes. The indium fraction in the InGaN dot alloy increases from the center to the edge of the wafer inversely proportional to the growth temperature.

The width of the dots varies from 20 to 40 nm and the height varies from 3 to 10 nm. The dots are completely embedded in the center of the nanowires. The nanowire is then capped with another 200 nm thick layer of GaN. The doping density estimated from the line width information from low temperature photoluminescence measurements[5] for this GaN nanowire sample (MN701) is about 10^{18} cm^{-3}.

The sample was optically characterized in the infrared region, specifically the Restrahlen region of GaN, using FTIR transmission spectroscopy. Measurements were done at the center, middle, and edge of the wafer with the edge having a higher nanowire density than the center of the wafer. Owing to the optical anisotropy[6] along the *c*-axis of the nanowires and the fairly consistent perpendicular orientation of the nanowires to the substrate surface, only transverse optical (TO) phonon modes can be observed with the longitudinal optical (LO) phonon modes almost entirely absent in the normal incidence transmission mode.[7] Characteristic surface optical (SO) modes of nanowires, which are longitudinal in nature, are also entirely absent. In order to study the TO, LO, and SO modes, we incorporated the elegant oblique angle incidence technique[6,8] in the transmission mode to exploit the Berreman effect.[9] Employing this technique, s and p polarized lights are made to be incident on the sample at 45° to the normal. With the s-polarized light (TE mode), the electric field component is parallel to the substrate and does

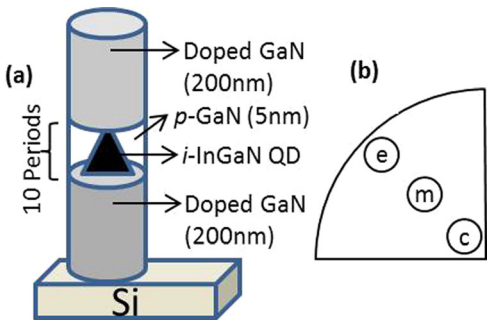

FIG. 1. (a) Schematic illustration of a dot-in-a-wire heterostructure on semi-insulating silicon substrate. (b) Schematic of a wafer sector indicating the spots where infrared transmission is done where "c" is center, "m" is middle, and "e" is edge of the wafer. The growth temperature decreases from the center to the edge of the wafer.

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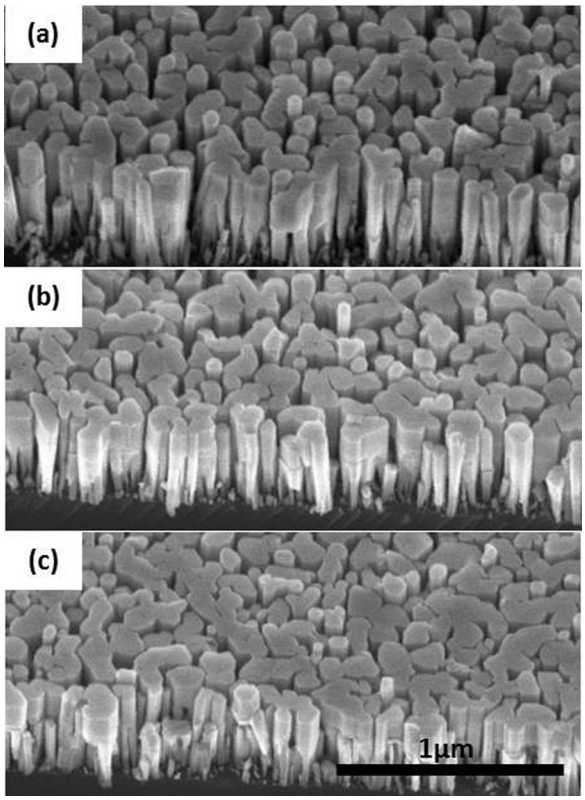

FIG. 2. (a) SEM image of a spot at the center of the wafer where the nanowire growth temperature is the highest and hence has the lowest nanowire fill factor. (b) SEM image of a spot at the middle of the wafer. (c) SEM image of a spot at the edge of the wafer where the nanowire growth temperature is the lowest and hence has the highest nanowire fill factor. Estimating from the area under the E_2 high TO phonon curve, the middle of the wafer has a filling factor ~1.5 times more than the center and the edge has a filling factor ~5 times more than the center.

not interact with the *c*-axis, hence only the TO mode is observed. But in the case of p-polarized light (TM mode), the electric field component is allowed to interact with the *c*-axis resulting in the observation of both TO and LO modes as shown in Figure 3. The observed TO mode at 559 cm^{-1} compares well with other published data for GaN nanowires.[10] However, the broad phonon modes in the sample (~705 cm^{-1}), which are longitudinal in nature, do not conform to any previously reported phonon modes for GaN.[11–13] The shift in the peaks toward the lower energy, as the nanowire density decreases, is characteristic of SO modes.[13] It is also observed that the SO mode evolves to the lower energy as the doping density of the nanowire is increased.

To understand the possible contribution of the InGaN dot to this anomalous phonon, the same measurements were conducted on a similar intrinsic GaN nanowire sample (MN478) but without the quantum dots. In addition to the (E_1)TO mode, similar SO modes were observed at ~650 cm^{-1}, which is also consistent with previously reported SO mode frequencies for GaN nanowires[11] as shown in Figure 4. This indicates that the shifted position of the SO modes in the sample (MN701) observed at the three spots on the wafer are due to the presence of the InGaN quantum dots embedded in the GaN nanowires which changes the dynamics of the surface lattice vibrations. In the doped sample (MN701), as the nanowire packing density is increased by moving the interrogation

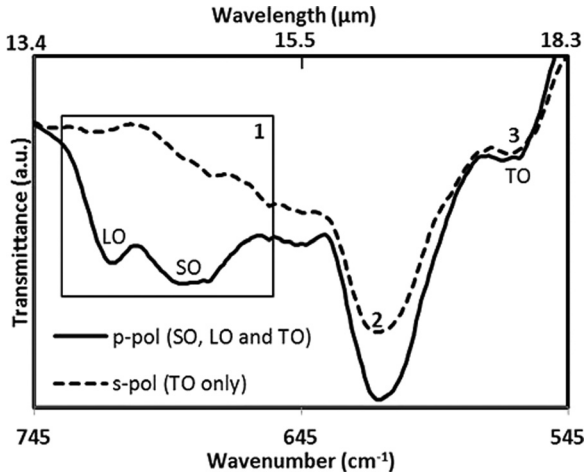

FIG. 3. Oblique angle transmittance performed using s- and p-polarized light on a doped (10^{18} cm^{-3}) GaN nanowire with embedded *i*-InGaN QDs (Sample MN701). The s-polarized light does not interact with the *c*-axis of the nanowire hence only TO (3) mode is evident. The electric field component of p-polarized light interacts with the *c*-axis resulting in SO, LO, and TO (1,3) modes. The broad transmittance dip at (2) is attributed to the vibrations of hydrogen bonded species,[14] C-C phonon, and substitutional carbon in the silicon substrate.[15]

spot from the center to the edge of the wafer, the SO modes evolve to the lower energy revealing a second peak which was eclipsed by the SO modes at lower nanowire density. This second peak is identified as LO phonon originating from the InGaN quantum dot. Further, considering the 20% indium fraction, the position of the LO peak is compatible with other published data.[16] The shifting of the LO mode of InGaN to the higher energy as the growth temperature is decreased from center to the edge of the wafer is due to the increasing indium fraction in the dots as a function of the temperature.

In order to ensure that these shifts are not due to LO phonon modes coupling with the plasmon wave, we conducted oblique angle transmission measurements at lower temperatures (250 to 20 K) by mounting the sample in a

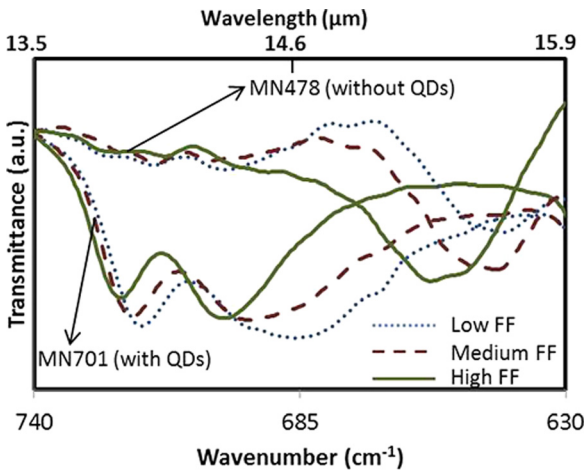

FIG. 4. An oblique angle transmission measurement done with p-polarized light. The suspected SO mode at ~705 cm^{-1} in sample MN701(with QDs) shifts to lower energies as the fill factor (FF) decreases. An LO mode is seen at ~722 cm^{-1}. The same measurement done on sample MN478 (undoped GaN without QDs) also indicates a shift of the SO mode (~650 cm^{-1}) to lower energies with decreasing fill factor, but no LO mode is seen indicating that the LO mode in MN701 is indeed due to the *i*-InGaN QDs.

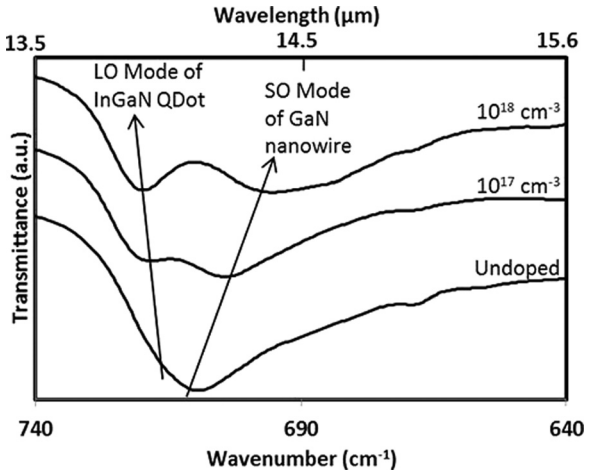

FIG. 5. Oblique angle measurement done on GaN nanowires with p-polarized light at varying doping densities with embedded i-InGaN QDs indicating a lower energy shift of the SO mode and a higher energy shift of the QD LO mode with increasing doping densities. In the undoped GaN nanowire sample, the LO mode of the QDs is eclipsed by the SO modes of the nanowires.

closed cycle refrigerated cold finger. The observed shift in the LO phonon was only about a couple of cm^{-1} wavenumbers hence ruling out the possibility of them having phonon-plasmon coupled origin. The observed shift in the position is more likely due to the radial breathing of the nanowires. Figure 5 shows the shifting of the LO mode arising from the InGaN dots shifting to the higher energy with higher doping densities. However, the full width at half maximum of the LO phonons remain constant which should not be the case of LO mode coupled with plasmon[17] which again verifies the absence of phonon-plasmon coupling.

Due to the polarization sensitivity of the SO phonon modes, we observe a high extinction ratio up to 72% between the s and p polarized light at oblique angle incidence. This would allow for the design of polarization sensitive infrared photodetectors.

In summary, we have reported the observance of surface optical modes by employing the oblique angle incidence technique, in InGaN/GaN dot-in-a-wire heterostructures whose positions are tunable by varying the nanowire density, the indium fraction of the dots, and the doping density of the nanowire. Moreover, the anisotropic nature of the *c*-axis of GaN offers polarization sensitivity. These properties of the dot-in-a-wire heterostructures make them a good candidate for phonon based infrared photodetectors.

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