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Resonant Raman scattering in self-assembled GaN/AlN quantum dots

Group of Spectroscopy of Solids

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N. Garro, A. Cros, J. M. Llorens, A. García-Cristóbal, and A. Cantarero
Institut de Ciència dels Materials, Universitat de València, Apdo. Correos 22085, E46071 València, Spain

N. Gogneau, E. Monroy, and B. Daudin
CEA-CNRS-UJF Research Group "Nonophysique et Semiconducteurs" Dépt. de la Recherche Fondamentale
sur la Matière Condensée, SP2M/PSC, CEA-Grenoble, 17 rue des Martyrs, 38054-Grenoble Cedex 9, France

Introduction

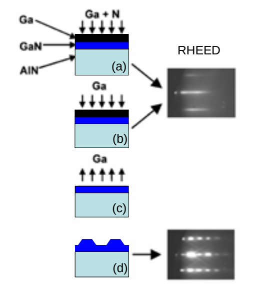
A considerable effort has been devoted in the last years to the achievement of self-assembled quantum dots (QDs). This interest is mainly motivated by their promising technological applications. Regarding optoelectronic devices, group III nitrides are the material support of emitters in the blue-ultraviolet wavelength range.

GaN/AlN heterostructures grown along the c-axis present an in-plane lattice mismatch that amounts to 2.4%. The resulting strain is partially relieved through the formation of 3D dislocation-free QDs that act as efficient room temperature emitters.

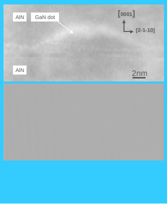
Optical properties of GaN QDs are determined by the interplay of confinement effects and the existence of strong built-in electric fields. Besides the spontaneous electrostatic polarization characteristic of III-V nitrides, piezoelectric fields induced by strain also contribute considerably to the internal electric field.

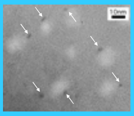
Raman scattering is an ideal probe of the strain field in semiconductor heterostructures.

Modified Stranski-Krastanow growth mode

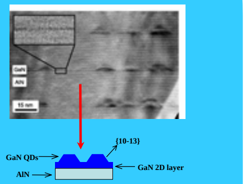


Schematic representation of the Modified Starsky-Krastanov growth Mode. In (a) GaN is grown under Ga rich conditions. The excess Ga adsorbs at the surface and forms a bilayer that stabilizes the two dimensional GaN layer below. (b) When growth is stopped under Ga rich conditions, the surface remains 2D. (c) Under vacuum, the Ga layer desorbs and the 2D GaN layer collapses to form 3D islands, shown in (d).





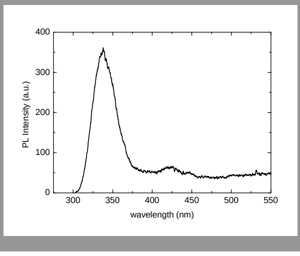
Threading edge dislocations (more than $10^{10}/cm^2$) are nucleation centers but GaN QDs are free of dislocations

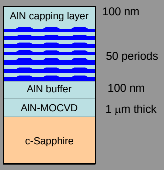


The QDs have the shape of a truncated pyramid, with a tilt angle of 32º, and grown on a 2 monolayers (ML) thick wetting layer (WL).

Sample description

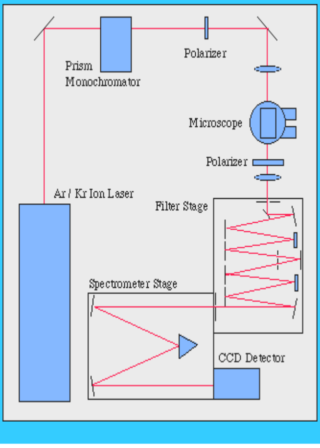
The sample studied was grown on a c-plane sapphire substrate in a MECA2000 plasma-assisted molecular beam epitaxy (PAMBE) chamber at a temperature of 750ºC, active nitrogen being supplied by a radio-frequency plasma cell. Its structure consists of stacks of 50 periods of GaN QDs separated by 8 nm thick AlN barriers. The nominal amount of GaN deposited is equivalent to 6 ML.





Room-temperature photoluminescence (PL) has been measured by exciting the structures with the fourth frequency of a pulsed Nd-YAG laser (wavelength = 266 nm, power density = 50 mW/cm²).

Confocal Micro-Raman Scattering Setup



Raman scattering was measured with a Jobin-Yvon T64000 triple spectrometer equipped with a confocal microscope. Several lines of an Ar+ laser were used as excitation. For the 514.5 nm laser line, the filter stage of the spectrometer has been avoided by the use of a Notch filter, enhancing considerable the Raman signal from the sample.

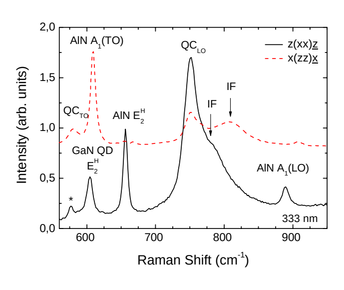
A 100x objective was used to focus the exciting beam on the sample for visible light, and a 40x for UV.

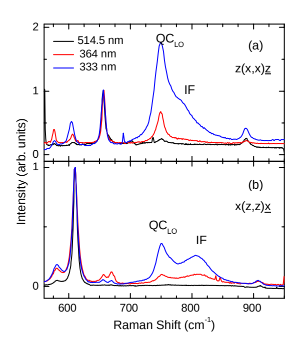
The holographic gratings of the system were changed from 1800 gr/mm to 2400 gr/mm to increase the resolution in the UV.

Resonant Raman scattering

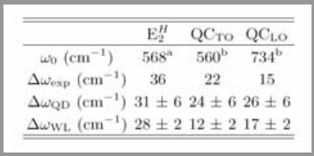
Raman spectrum shows the vibrational modes of the QDs and the AlN barriers. While the phonons of the AlN spacers appear at their bulk frequencies, the modes of the QDs are considerably blue-shifted. Thus the AlN barrier is almost fully relaxed. Raman modes of the QDs, on the other hand, are shifted by the strain field. Besides the strain effects, polar vibrational modes manifest confinement effects.

- Quasi-confined (QC) polar modes are observed at different polarization configurations.
- Interface (IF) polar modes appear at higher energies and exhibit large linewidths, as predicted by recent theoretical calculations.





Strain-induced Raman frequency shifts are summarised in the table, together with the results of the theoretical model for the QD and the biaxially strained WL. The experimental frequency shifts of the QDs lay far from the biaxial limit, except for the QC_{LO} Raman peak. The intensity of this peak is dominated by Fröhlich interaction with excitons partly localized in the WL underneath the QDs.



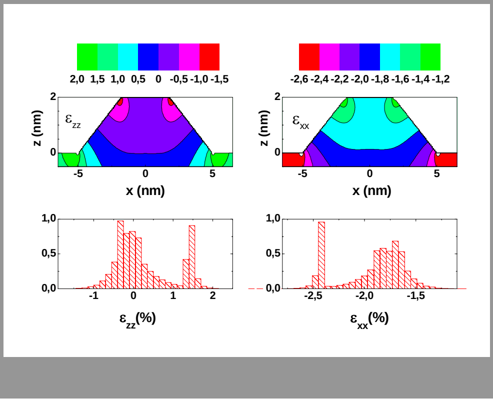
	E_2^H	QC_{TO}	QC_{LO}
ω_0 (cm^{-1})	568^a	560^b	734^b
$\Delta\omega_{exp}$ (cm^{-1})	36	22	15
$\Delta\omega_{QD}$ (cm^{-1})	31 ± 6	24 ± 6	26 ± 6
$\Delta\omega_{WL}$ (cm^{-1})	28 ± 2	12 ± 2	17 ± 2

There is a resonant enhancement of Raman intensities of the QC and IF modes when resonant conditions are approached. This proves the polar character of these modes and their adscription to the QDs.

Theoretical model

The strain distribution in truncated cone QD, the 2D WL, and the AlN barrier around them has been calculated with a theoretical model based on the method of inclusions. The phonon frequency shift induced by strain is given by

$$\Delta\omega_\lambda = 2a_\lambda \varepsilon_{xx} + b_\lambda \varepsilon_{zz}$$



Conclusions

- Quasi-confined and interface modes have been observed in the Raman spectra of self-assembled GaN/AlN quantum dots.
- The broadband character predicted for interface modes induced the large linewidths of the Raman peaks.
- Additional Raman shifts can be attributed to the strain field.
- Good agreement is found between the experimental and theoretical results, which proves the elastic character of the strain relaxation in the quantum dots and its deviation from the 2D biaxial strain.

References

- N. Gogneau, D. Jalabert, E. Monroy, T. Shibata, M. Tanaka, and B. Daudin, J. Appl. Phys. **94,** 2254 (2003).
- N. Garro, A.Cros, J. M. Llorens, A. García-Cristóbal, A. Cantarero, N. Gogneau, E. Monroy , and B. Daudin, submitted.
- D. A. Romanov, V. V. Mitin, and M. A. Stroscio, Phys. Rev. B **66**, 115321 (2002).
- A. D. Andreev, and E. P. O'Reilly, Phys. Rev. B **62**, 15851 (2000).