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Report:

GaAsN and InGaAsN are peculiar alloys among the III-V family from an optical and electronic standpoint since an anomalously large and composition dependent optical bowing has been recently reported. It is by now accepted that this effect is basically due to the large difference in size, orbital energies, and electronegativity between As and the substitutional N atoms. The giant bowing induces a red-shift of the band gap as large as 0.1-0.2 eV per N percent, which enables achieving (in the case of InGaAsN) wavelengths of great interest in fiber-optics telecommunications and renewable energy (λ = 1.3-1.55 μm). Multi-junction solar cells that implement this new technology are under development while InGaAsN quantum wells are nowadays employed as active layers in Vertical Cavity Surface Emitting Lasers (VCSEL), exploited in fiber- optic networks at the metro and local area. Technological interest has triggered studies on the fundamental physics of III-V dilute nitrides, in particular on the optical and electronic properties; on the other hand, little information is available on the structure of these alloys, in particular at the local microscopic scale.

In order to study the variations in the local structure of the GaAs host lattice upon incorporation of nitrogen we have performed X-ray absorption (XAS) measurements at the Ga K edge on the GILDA beamline of ESRF. The experiment was performed using a dynamical sagittaly focusing Si (311) monochromator and higher order harmonics were rejected by exploiting two Pd-covered mirrors and detuning the crystals. The energy dependence of the X-ray absorption coefficient at the Ga K- edge was recorded by measuring the drain current from the sample (total electron yield, TEY). We measured the three GaAsN epilayers plus a GaAs standard, in the same experimental conditions; the GaAsN epilayers were 300 nm thick with N concentration up to 2.68%. TEY detection allows to selectively probe only the epilayers, whose thickness is 300 nm. In fact even though KLL Auger electrons are quite energetic, based on a total-electron-yield depth dependence model we estimate the contribution of absorption events taking place in the substrate to the total electron current to be lower than 3 - 4% in our experiment. The flux on the sample was $3*10^{10}$ photons/s in a spot of 1 mm^2. In Fig. 1 we report the background – subtracted EXAFS spectra, illustrating the high signal – to – noise ratio obtained.

Data analysis was performed by *ab initio* modeling of the absorption cross section by the FEFF 8.00 code, the theoretical backscattering amplitudes and phase shifts were calculated using cubic GaAs and GaN model clusters. The Feffit program was exploited to fit data in R-space within the window [0 - 4.9] Å which includes the second and third coordination shells. We were not directly sensitive to the N coordination in the first shell, because of the high N dilution and of low backscattering amplitude of nitrogen atoms; our goal was to measure effects induced by the presence of nitrogen on the average Ga-As bond lengths and their distribution.

Data analysis indicates that incorporation of N induces a small but detectable increase of the average Ga – As bond length and an increase in the width of the interatomic distance distribution (as measured by the EXAFS Debye – Waller factor). The increase in the Ga – As bond length can be understood as due to the competing effects of the decrease of the free lattice parameter induced by N and the tensile strain due to the pseudomorphic growth on GaAs. Fig. 2 we report the comparison between the experimental Ga – As bond length and the predictions of various theoretical estimates: three valence force field models (by: Cai and Thorpe, CT; Martins and Zunger, MZ; Balzarotti et. al., B) and original Density Functional Theory simulations; the VFF models have been modified to take into account the effect of strain. While good agreement is found for the DFT calculations and for model B, the predictions of CT are outside the 2σ limit. We believe that this is due to the fact that model CT does not take into account the differences in the bond bending and bond stretching force constants, which is not negligible in GaAsN due to the very different size and electronegativity of As and N.

In Fig. 3 we report the comparison between the experimental Debye – Waller factors and those predicted by the DFT simulations; a good agreement can be noticed. The increase in the DW factor can be understood as a consequence of the static disorder induced on the Ga – As bond by the strong Ga – N interaction.

The results of this experiment are reported in more detail in:

G. Ciatto, F. D'Acapito, S. Sanna, V. Fiorentini, A. Polimeni, M. Capizzi, S. Mobilio and F. Boscherini, accepted for publication in Physical Review B (to be published in March 2005).

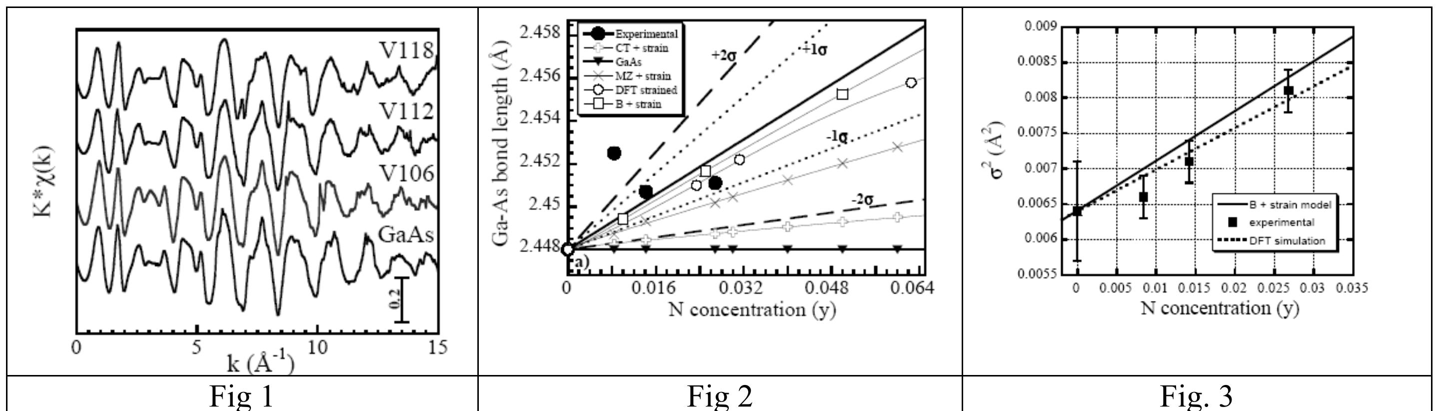

Fig 1	Fig 2	Fig. 3