Interface formation of scandium nitride on GaN(0001) surface: A first-principles study

Rafael González-Hernández[1], William López-Pérez[1], Jairo Arbey Rodríguez M.[2] Aldo H. Romero[3]

[1]Department of Physics. Universidad del Norte. Km 5 vía a Puerto Colombia, A. A. 1569. Barranquilla, Colombia.

[2]Department of Physics. Universidad Nacional de Colombia. A. A. 5997, Bogotá, Colombia.

[3]CINVESTAV, Departamento de Materiales, Unidad Querétaro, Querétaro, 76230, México

e-mail: rhernandezj@uninorte.edu.co

We have carried out first-principles calculations in order to study the energetic and electronic structure of scandium (Sc) adsorption, diffusion and incorporation on a GaN(0001) surface using density-functional theory (DFT) within a plane-wave ultrasoft pseudopotential scheme. The calculations are based on an eight-layer slab of wurtzite 2x2-GaN(0001) supercell, with 11 Å of vacuum as the spacing layer in the growth direction. The dangling bonds at the bottom N-layer were saturated with pseudohydrogen atoms with a fractional charge of 0.75*e*. The upper two bilayers of GaN and adatoms were allowed to relax, while the bottom two bilayers of GaN were fixed at their bulk positions to mimic the bulk substrate.

The results show that Sc atoms preferentially adsorb at the T_4 sites at low and high coverages (from 1/4 up to 1 monolayer). It is also found that N atom is relatively immobile (surface energy barrier of 1.4 eV) compared to Ga (0.5 eV) as well as Sc (0.7 eV) atoms on the GaN(0001) surface. In addition, calculating the relative surface energy of several configurations and various Sc concentrations, we constructed a phase diagram showing the energetically most stable surfaces as a function of the Ga chemical potential. Based on these results, we found that incorporation of Sc adatoms in the Ga-substitutional site is energetically more favorable compared with the adsorption on the top layers. This effect leads to the formation of an interfacial crystalline ScN layer on the GaN(0001) surface, which can offer a good interfacial combination between Sc and GaN substrate. Our calculations show that the scandium incorporation is most favorable under a nitrogen environment, in agreement with experimental results.

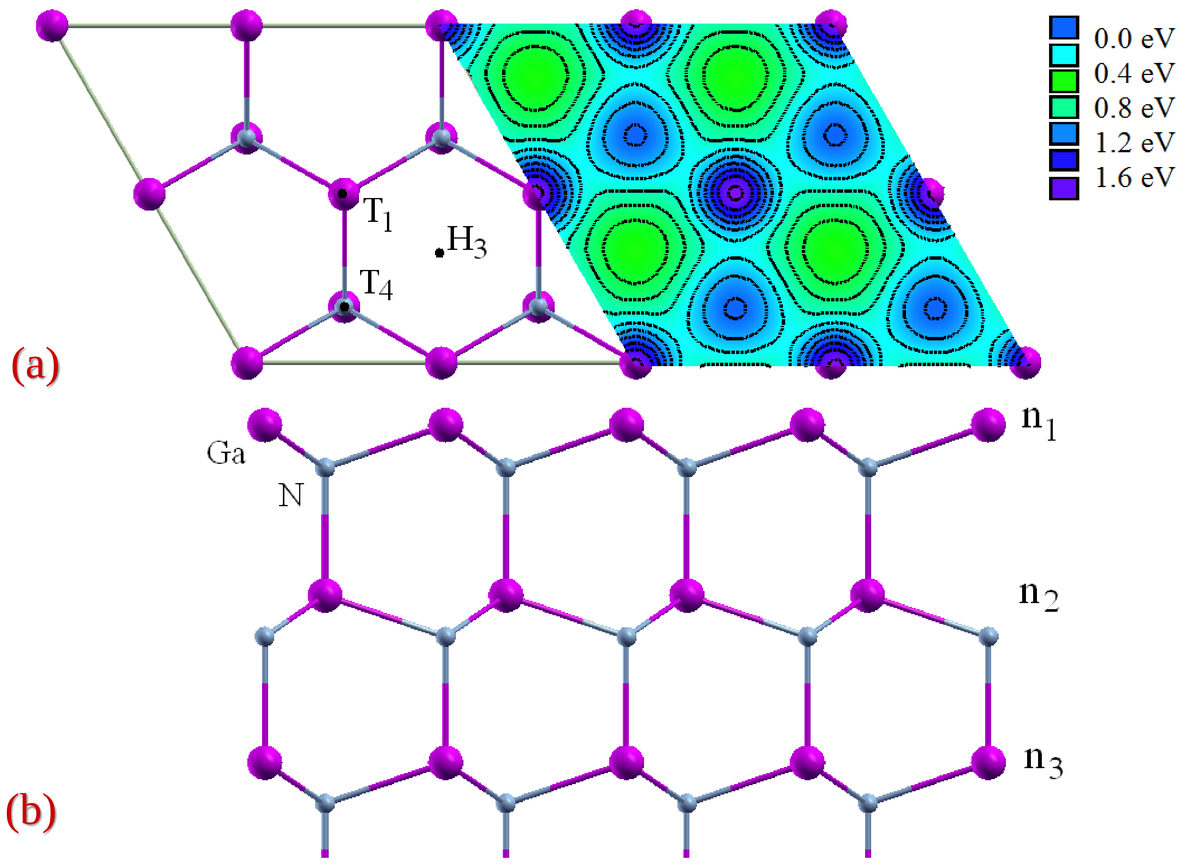

Fig. 1. (a) Top view of the 2x2-GaN(0001) surface with the high symmetry adsorption sites indicated and contour plot of PES for Sc adatom diffusion. (b) Side view of the 2x2-GaN(0001) unit cell. The values ($n_1/n_2/n_3$ /) indicate the number of Sc impurities in the Ga-layer.

The surface stoichiometry is not the same for all the studied configurations, the formation energies depend upon the chemical potential of the atomic species in excess. The relative formation energy E_f is:

$$E_{formation}=E_{total}-E_{ref}-\Delta n_{Sc}\mu_{Sc}-\Delta n_{Ga}\mu_{Ga}-\Delta n_{N}\mu_{N}$$

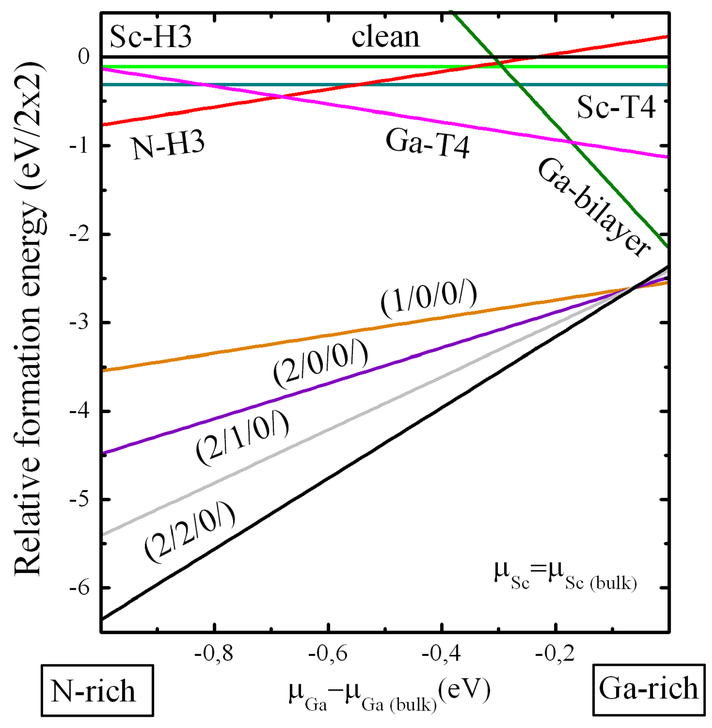

Fig. 3. Relative formation energies of various Sc impurity configurations as a function of the relative chemical potential of Ga. The zero of the energy scale corresponds to the formation energy of the clean GaN(0001)-2x2 surface. The notation is introduced in Fig.1

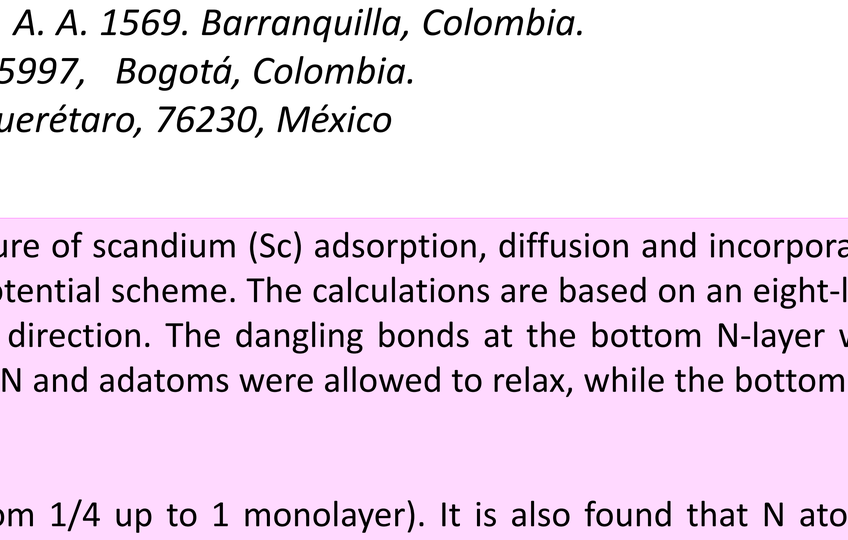

Fig. 2. Formation energy of different configurations relative to the clean 2x2-GaN(0001) surface.

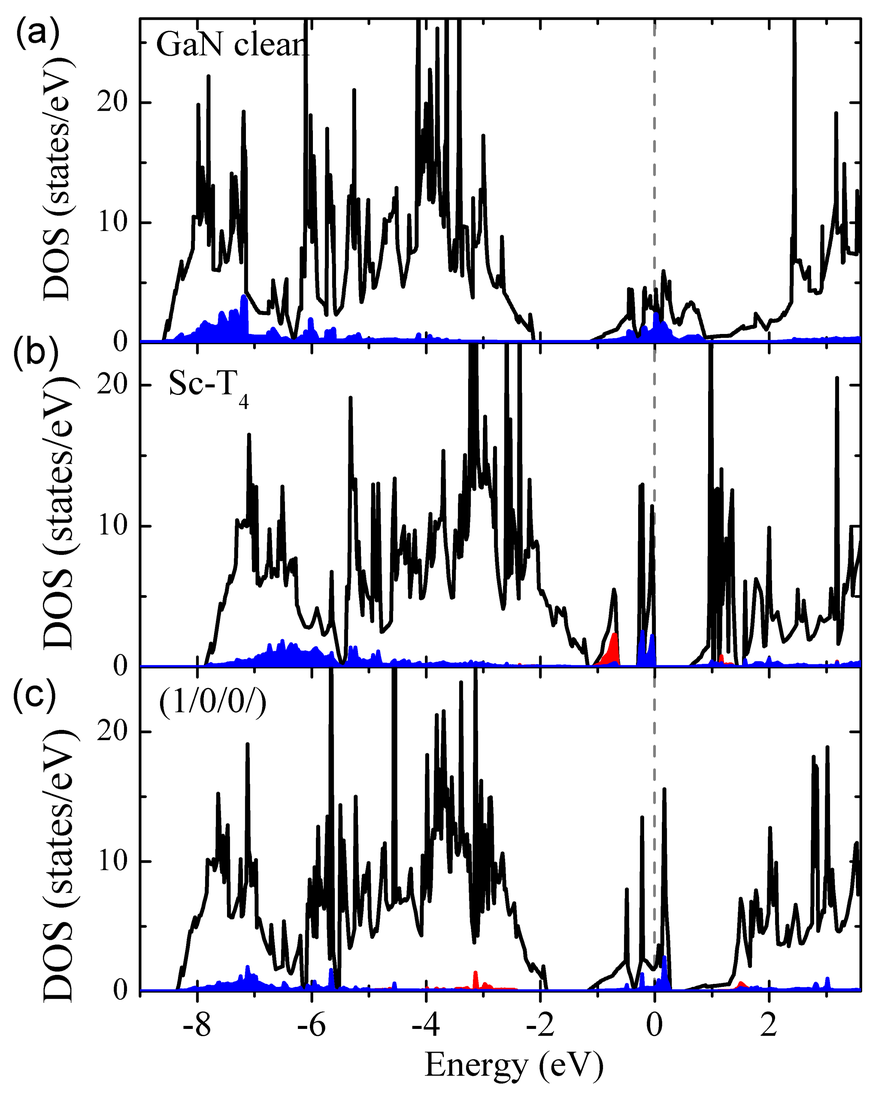

Fig. 4 (a) Total density of states (DOS) for the relaxed GaN(0001) clean surface. Ga-top layer projected DOS is shown (blue area). (b) Total DOS for the Sc-T4 GaN(0001) surface reconstruction. (c) Total DOS for the (1/0/0/) configuration. Sc-impurity and Ga-top layer projected DOS are shown (red and blue area, respectively). The Fermi level is to zero *eV*.