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### Molecular beam epitaxial growth of 2D-boron nitride on Ni substrates

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Abstract – 2D boron nitride (2D-BN) was synthesized by gas-source molecular beam epitaxy on Ni substrates using gaseous borazine and active nitrogen generated by a remote plasma source. Based on spatially resolved photoemission spectroscopy and on the detection of the  $\pi$  plasmon peak, we discuss the origin of the two N and B main components and their relationship with an electronic coupling with Ni at the interface. After optimization of the growth parameters, a full 2D-BN coverage is obtained although the material thickness is not homogeneous, showing triangular islands whose lateral size is limited to ~20 µm.

#### Keywords-boron nitride, borazine, epitaxy, plasma-assisted

The growth of 2D-BN by gas source molecular beam epitaxy was studied and optimized, using the borazine precursor associated with activated nitrogen as provided by a remote valve plasma cell. The goal was to grow 2D-BN on Ni polycrystalline foils under high-vacuum conditions, this peculiar set of precursors being selected in order to overcome the thickness self-limitation observed when using borazine alone<sup>1</sup>.

A typical X-ray photoemission N1s spectrum is displayed in Fig. 1 after optimization of the main growth parameters, showing two components shifted by ~1 eV as well as the  $\pi$  plasmon peak associated to 2D-BN<sup>2</sup>. These two components are respectively associated to the interface BN layer exhibiting a strong hybridization between Ni and BN states<sup>3</sup> and to the other top BN layer(s) bound by Van der Waals interactions. Although the 2D-BN fully covers the substrate, the growth does not follow a layerby-layer mode. Instead, triangular multilayer domains are observed as depicted in Fig. 2, with a typical maximum dimension of 20 µm. These domains involve stacks of 2D-BN, up to 15 ML thick, showing high-quality material characteristics. The Raman spectrum shown in Fig. 3a has a full width at half maximum of 10-11 cm<sup>-1</sup>. The cathodoluminescence spectrum displayed in Fig. 3b shows a peak at 6.045 eV, associated to the non-thermalized recombination of direct excitons in hBN<sup>4</sup>. The valence band structure measured by nano-angle resolved photoemission spectroscopy along the  $\Gamma K$  direction (see Fig. 3c) is the one expected for 2D-BN. Conducting atomic-force microscopy imaging and a corresponding current profile are displayed in Fig. 4. The current perpendicular to the BN stack strongly decreases when the thickness increases, being no longer measurable (below 1 pA for a bias voltage of 1 V) from 5 ML and thicker 2D-BN. All these information confirm that high quality 2D-BN insulating material can be grown by combining borazine and activated nitrogen, although homogeneous thick layers are not yet obtained.

<sup>&</sup>lt;sup>1</sup> Hadid et al. 2D Materials 8, 045007 (2021)

<sup>&</sup>lt;sup>2</sup> Trehan et al. J. Vac. Sci. Technol. A 8, 4026 (1990)

<sup>&</sup>lt;sup>3</sup> Preobrajenski et al. Phys. Rev. B 70, 165404 (2004)

<sup>&</sup>lt;sup>4</sup> Schué et al. Nanoscale 8, 6986 (2016)

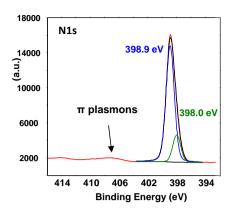


Figure 1:. A typical XPS N1s spectrum showing two main N1s components as well as the  $\pi$  plasmon peak.

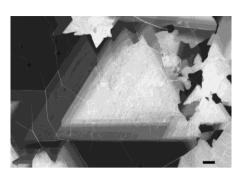
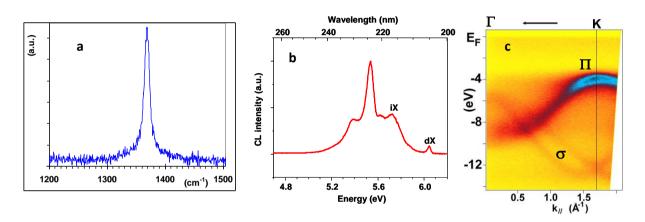
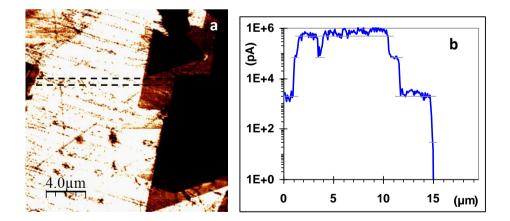


Figure 2:. A large BN triangular domain, up to 15 BN layer thick (scale bar 2  $\mu$ m).



*Figure 3*: optimized BN growth at 910°C, triangular domains, a) typical Raman spectrum (FWHM 10-11 cm<sup>-1</sup>), b) 5 K cathodoluminescence spectrum and c) nano-ARPES valence band structure (photon energy 100 eV).



*Figure 4*: optimized BN growth at 910°C, triangular domains, a) conductive AFM image and b) average current profile measured in the area delimited by the dashed lines in (a).