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Letter to the Editor

Reversed texture in nanometric carbon/boron nitride multilayers

R. Torres [a,b], I. Caretti [a], V. Serin [b,*], N. Brun [c], G. Radnóczic [d], I. Jiménez [a]

[a] ICMM-CSIC, Campus de Cantoblanco, 28049 Madrid, Spain
[b] CEMES, Université Toulouse, 29 rue Jeanne Marvig, BP 94347, 31055 Toulouse, France
[c] LPS, UMR 8502 CNRS, Université Paris Sud, Bât. 510, Orsay F-91405, France
[d] Centre for Natural Sciences, ITPMS, Konkoly-Thege M. út 29-33, 1121 Budapest, Hungary

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ABSTRACT

A structure-controlled series of carbon/boron nitride multilayers, with bilayer thicknesses from 1.25 to 160 nm has been grown by sequential evaporation of carbon and boron assisted with nitrogen ions. The minimum bilayer thickness for a stable stack is 2.9 nm. A turbostratic texture of the carbon and BN phases is evidenced even for small periods of the bilayers. Interestingly, BN and C basal planes of adjacent sub-layers exhibit perpendicular alignment between them: along the growth direction for h-BN rich layers, and parallel to the surface for the C rich ones.

In this work we have faced up to the objective of growing carbon/BN multilayers by physical vapor deposition. Since both components present hexagonal structure with similar lattice parameters, it is interesting to study the formation of ordered super-lattices or, on the contrary, if distinct orientations are formed in each phase. In addition, if non-abrupt, gradual interfaces occur, the carbon/BN system keeps open the possibility of forming ternary BCN compounds. Highly ordered carbon/BN with parallel stacking have already been synthetized with regards to basic graphene research by exfoliation-transferring methods [1,2], that are unpractical for real applications. Therefore, there is a renewed interest in combined carbon/BN growth methods with large throughputs. The carbon/h-BN multilayers were obtained by the sequential ion-beam assisted-deposition (IBAD) of carbon and boron nitride in a vacuum chamber with a configuration described in [3] and sketched in Fig. 1.

Two electron guns were used to evaporate independently carbon and boron, and an ion gun directed towards the boron evaporation position provided the supply of nitrogen ions. As substrates we used pieces of Si (100) wafers, mounted on a turntable that passed sequentially through the C and the B–N fluxes. As solid precursors for the vapors we used carbon cylinders with 99% purity (Alfa-Aesar 10134) and boron pieces with 99.5% purity (Alfa-Aesar 12134). The evaporation was carried out using two electron beam evaporators operating at 7 kV, with the emission currents adjusted to achieve a C:BN thickness ratio equal to 1. The ion assistance was carried out with a 3 cm diameter Kauffman ion gun fed with 5 sccm of N_2 (99.9995% purity), supplying 500 eV ions. The base pressure was 1×10^{-7} mbar and the working pressure during deposition was 1×10^{-4} mbar. The substrates were not heated intentionally, but their temperature during growth reached ~200 °C, due to ion stopping and radiation from the

* Corresponding author.
E-mail address: serin@cemes.fr (V. Serin).
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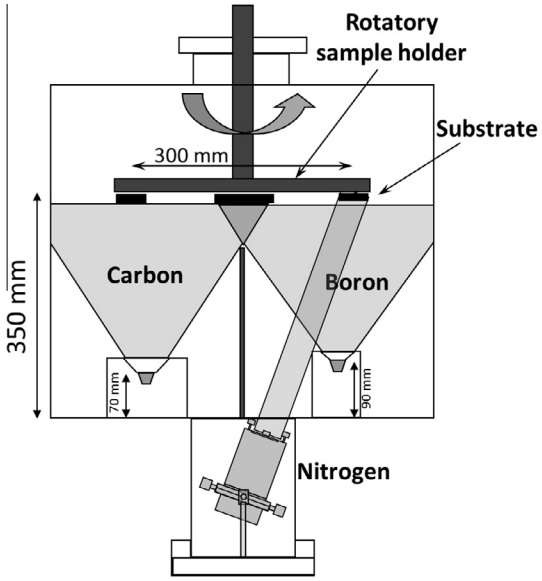

Fig. 1 – Scheme of the IBAD-PVD deposition system.

evaporators. With the deposition conditions used, the bonding structure for both materials is hexagonal. A series of multilayers were prepared using the same evaporation conditions but doubling the rotating speed of the turntable for each new sample to obtain half the period. Using this procedure we prepared a set of multilayers with an expected bilayer or period length of 160, 80, 40, 20, 10, 5, 2.5 and 1.25 nm. These expected values are denoted as "nominal period" and have been found experimentally to vary along the multilayer thickness due to problems with the stability of deposition rates. For the sake of clarity, we will denote as multilayer the whole film, which is composed of C/BN bilayers, and will use the term sublayer to refer to each individual C or BN layer.

The multilayer's micro and nanostructure were investigated by means of transmission electron microscopy (TEM) and Electron Energy Loss spectroscopy (EELS) performed on an image-corrected FEI Tecnai operating at 200 kV and equipped with a Gatan imaging filter. Energy filtered images were acquired using a JEOL 3010 operating at 300 kV equipped with a TRIDIEM EELS spectrometer.

Cross-sectional TEM images of several samples with different period lengths have been employed to take side views of the multilayers. The samples were very stable under the beam, any damage has been observed. Fig. 2 displays images of some multilayers.

Alternated contrast bands, detected from the substrate to the surface of the multilayers, confirm that the sequential deposition of C and BN results in a stable multilayer structure. The unexpected difference of TEM contrast between the sublayers, made of elements with such close Z numbers, is related to thickness effects due to the TEM preparation, as has been checked by recording EELS thickness maps. From the images, two effects are noticed. The first one is the shortening of the bilayer length while moving from the substrate to the surface. This is a technical effect caused by the decrease of the evaporation rate from the source as the material is consumed, and can be easily compensated with a feedback loop. The second effect is the increase of interfacial roughness,

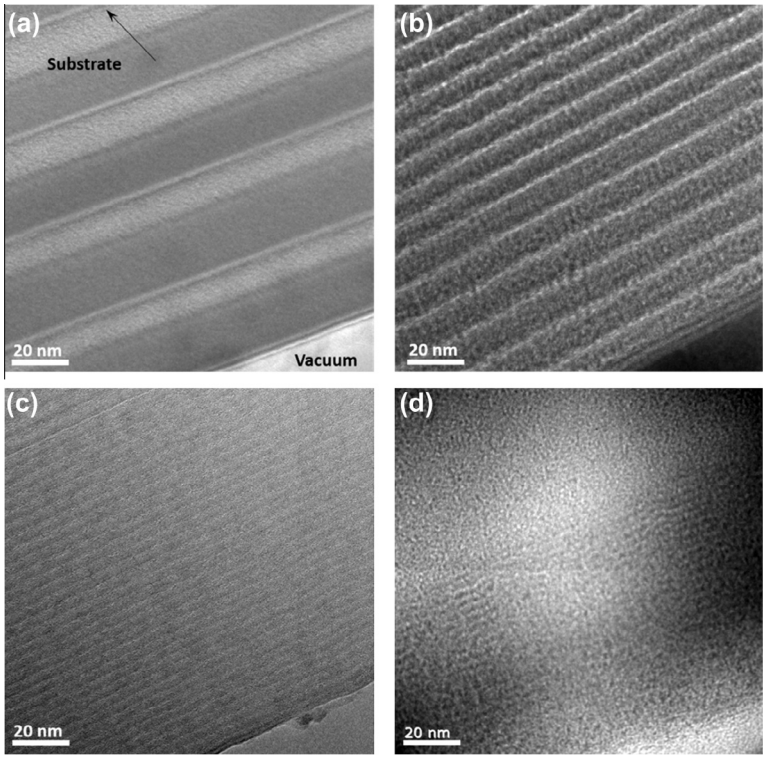

Fig. 2 – Low magnification images of C/BN multilayers with nominal multilayer period of (a) 40 nm, (b) 10 nm, and (c) 5 nm, (d) 2.5 nm.