Deposition of ultra-thin graphene-like coatings by PECVD methods

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Abstract

Since its first theoretical descriptions in the 50's of the last century and experimental evidence in 2005 a large number of production processes for graphene have been proposed. However, only a small part of them is interesting in terms of up-scaling into a cheap and reliable industrial production. Plasma enhanced chemical vapor deposition (PECVD) offers this potential but only few reports about successful synthesis exist [1,2]. This contribution presents some results of thin and ultra-thin diamond-like carbon coatings (DLC), deposited and modified by PECVD with the aim to produce graphene-like structures on top of Si and Si/SiO_2 wafers.

The coatings were deposited on standard single side polished <100>, P/Bor doped, 525±25 µm thick Si wafers, with and without 100 nm thermal SiO_2 layer. Prior to deposition the wafer were cleaned chemically in an ultrasonic cleaner with aceton and ethanol and then by argon plasma (nominal purity >99.999%), using an ALS340L anode layer source from Veeco Instruments (Woodbury, NY, USA) with a rotating carrousel, at 3 kV accelerating voltage and 30 sccm gas flow rate. The coatings were deposited by employing the same ion source with 20 sccm C_2H_2 (nominal purity >99.96%) and 1 and 3 kV voltages and subsequently etched in Ar plasma at 1 and 3 kV. Deposition times were varied within 1.5 to 60 minutes, etching times from 30 seconds to 40 minutes. Several coatings were deposited statically at deposition and etching temperatures of 400°C and reduced deposition and etching times (5 – 15 seconds). Some coatings were tempered in a nitrogen-filled tubular furnace at 300° and 800°C for 8 hours, 15 minutes, respectively.

The coating thickness, measured by profilometry and ellipsometry, ranges between 10 and 92 nm. The measured optical constants of the coatings, namely n (refraction index, Fig. 1) and k (absorption coefficient) in the range 300 - 1000 nm, are most similar to the characteristics of graphite and graphene [3,4] when deposited at 3 kV for 60 minutes, and ion etched at 800 eV for 40 minutes.

Raman spectroscopy of the as-deposited and the etched and tempered DLC coatings revealed a slight increase in the wavenumber of the G-band and the intensity ratio of the D- and G-band I_D/I_G, accompanied by a decrease in full width at half maximum (FWHM) of the G-band (Fig. 2). The effect was much more pronounced for the coatings deposited at higher temperatures and the tempered coatings. Such spectral changes can be interpreted as transformation from DLC coatings to nanocrystalline graphite, accompanied by increasing structural order and crystallite size of sp^2 coordinated carbon [5]. The smaller effect of plasma treatment might result from the relatively high ion etching energies of 800 to 1500 eV, increasing rather the structural disorder and crystallite size of the coatings instead of selectively removing comparable lower stable sp^3 coordinated carbon, as it was expected.

References

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Figures

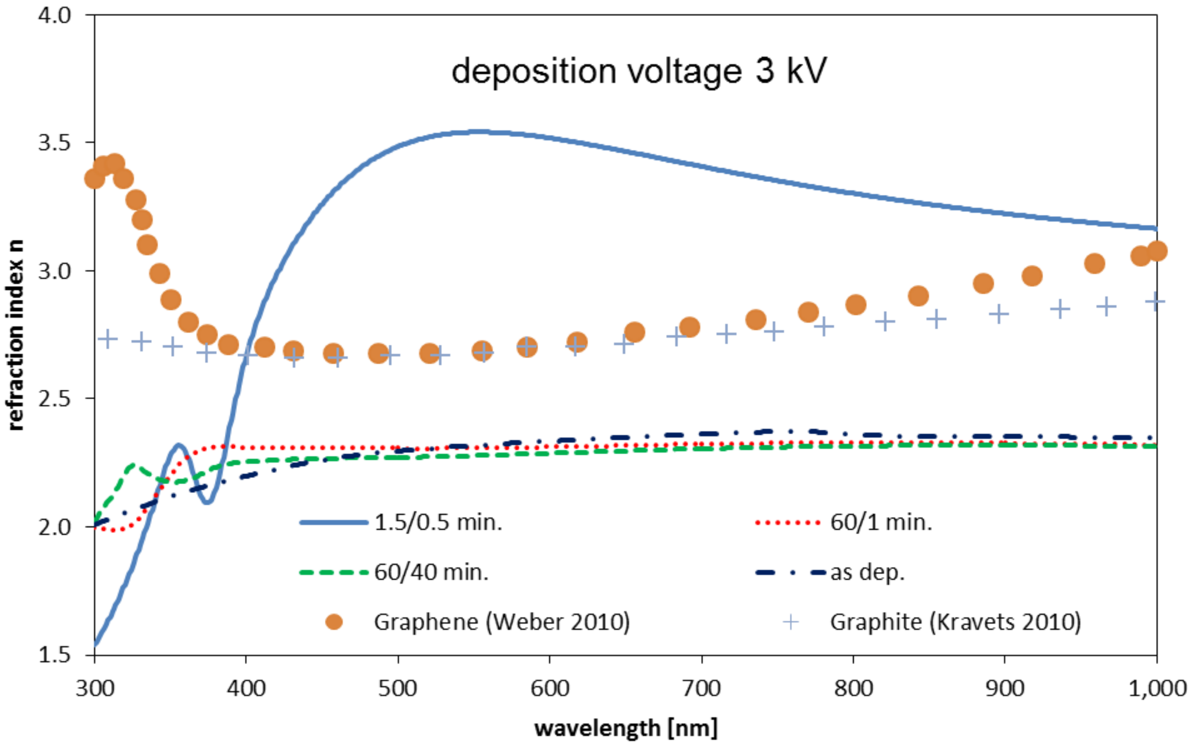

Figure 1: Refraction index n of three plasma-etched DLC coatings (solid, dotted, dashed lines), legend indicates deposition/etching time. Chain-dotted line shows as deposited DLC coating. Graphene and graphite are represented by circles and crosses.

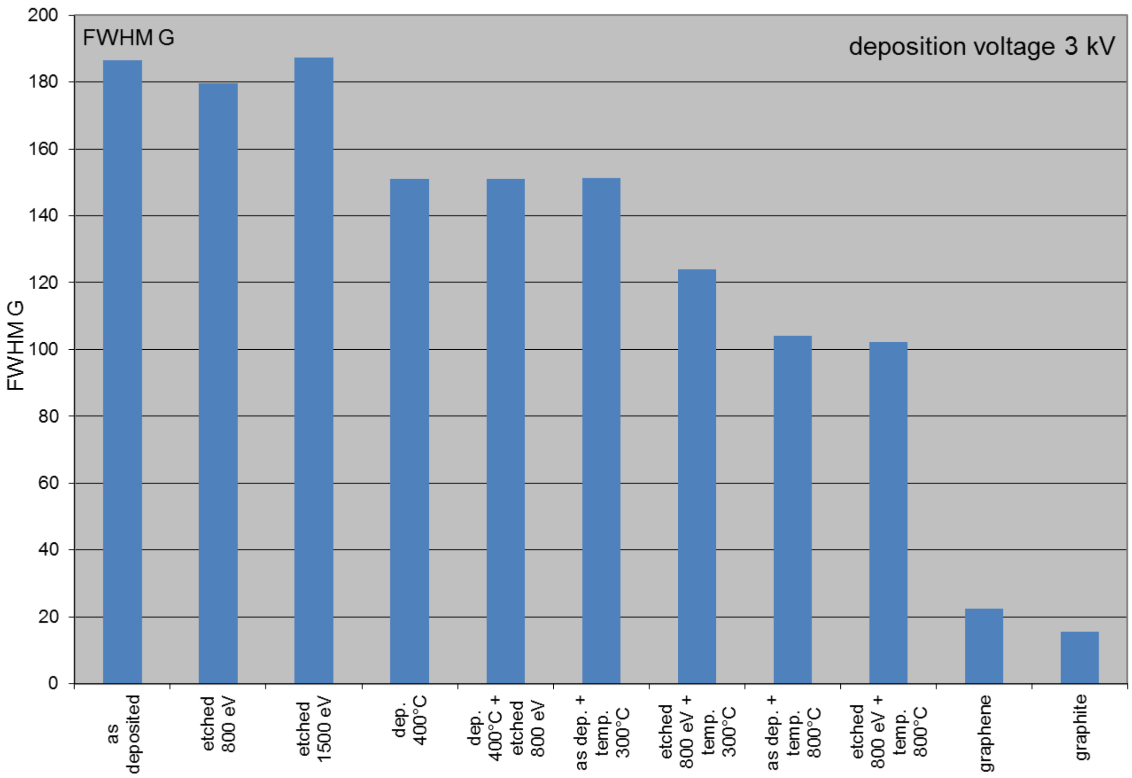

Figure 2: Full width at half maximum of the Raman G-band (FWHM G) of DLC coatings, deposited at 3 kV accelerating voltage, at various deposition and tempering temperatures and ion etching energies. FWHM G of graphene and graphite are given for comparison.