Supplementary Material for the manuscript:

Characterization, Optimization, and Surface Physics Aspects of in-situ Plasma Mirror Cleaning

For the deposition of carbon layers onto the various test objects described in the main article a commercial e-beam deposition chamber has been used, that allowed for a deposition of about 100 nm of carbon within 120 seconds. A comparative C1s XPS analysis of the resulting carbon coating with (i) the graphitic target material as well as (ii) with highly oriented pyrolytic graphite (HOPG) was performed in order to shed some light on the differences in the electronic structure of these materials. Fig. 1 shows the resulting C1s XPS spectra. The larger FWHM of 1.6 eV for the C1s line of the carbon thin film as compared to the FWHM of 0.6 eV for both the HOPG as well as the graphitic carbon target material indicate an increase of C atoms with sp^3 configuration in addition to the already existing C atoms in sp^2 configuration and thus an amorphous character for the former C thin films.[1]

The test chamber setup for the plasma cleaning did consist of a custom UHV chamber pumped by a corrosion-resistant combination of a main turbo pump, a turbo drag backing pump, and a diaphragm pump in order to cover a wide range of gas pressures utilizing either a static or a dynamic gas supply regime for the cleaning process.

In the case of static gas supply conditions, the constituents of the plasma inside the cleaning chamber were analysed using a differentially pumped residual gas analyser (RGA), again for covering a wide range of plasma operation gas pressures while maintaining the RGA below its upper operation pressure limit during the cleaning process via an adjustable orifice for the differential pumping. However, it has to be noted that an exhaustive observation of plasma gas species is only possible in the case of static gas supply conditions where all gas educts and products are conserved inside the chamber volume and thus available for RGA observation as a function of process time. Also, due to the well-known gas de-mixing phenomena at small orifices[2] such as the 0.1 mm entrance orifice of the differentially pumped RGA chamber, measured RGA mass ratios of gases with highly different atomic masses and viscosities have to be assessed carefully when it comes to a quantitative evaluation of the mass spectra.

Graphitic carbon cleaning rates were determined using a commercial quartz crystal microbalance (SQM-160, Inficon Inc., East Syracuse, NY, USA) inside the cleaning chamber equipped with the corresponding quartz balance crystals that were previously coated with a graphitic carbon layer using the e-beam deposition chamber mentioned above.

Optical UV/VIS spectra of the plasma emission were taken using commercial grating spectrometers, either with a wide wavelength range and low resolution or narrow wavelength range and high

resolution. The analysis of the plasma optical emission spectra and the peak assignment was performed using the SpecLine software.[3]

Gas mixtures were prepared using a custom-made gas mixing system, with the gas ratios determined either by partial pressures read off from either a Pirani or diaphragm gauge, or by comparing the intensity of optical emission lines from the different gases.

Subsequent to the cleaning run, the noble metal foil test samples were analysed for changes regarding their surface chemistry by XPS using a (monochromatized) Al Kα x-ray source together with a SPECS Phoibos 150 electron analyser set to an electron pass energy of 25 eV (medium area mode, 20 $\times$ 7 mm^2 analyser entrance slit) resulting into an overall electron kinetic energy resolution of about 0.5 eV.

Changes in surface morphology were analysed using the Au-, Rh-, or Ni-plated single crystalline Si substrates polished to optical quality mentioned above (see Fig. 3(d)) together with a Veeco/Wyko NT9300 optical profilometer.

Fig. 2 shows the in-UHV aluminum antennae used in the context of the traditional capacitive coupled plasma sources (CCP) as well as the inductively coupled plasma gun (ICP). The chemical and kinetic interaction of the plasma with the surface of the aluminum CCP antenna is obvious from the colored oxide traces on the surface of the cylindrical antenna body.

Fig. 3 shows the various test objects used in the context of the quantitative analysis of the plasma cleaning. Initial tests were performed using the carbon-coated aluminum disks shown in (a), whereas the quartz crystal microbalance (QCM) crystals shown in (b) are used for the quantitative determination of the carbon cleaning rate. The metal foils mounted inside the foil holder shown in (c) were used for the XPS analysis of the chemical interaction of the plasma with the metal surfaces. The partially carbon coated test mirrors shown in (d) are used for determining changes in the surface morphology and micro-roughness of their optical surfaces.

Tables 1 and 2 give a compilation of the results from the micro-roughness measurements on the test mirrors shown in Fig. 3(d) using an interference microscope. Table 1 shows the micro-roughness of the test mirrors after (partial) carbon-coating and subsequent O_2/Ar plasma cleaning. Table 2 shows the micro-roughness of the test mirrors after (partial) carbon-coating and subsequent H_2/Ar plasma cleaning. At a glance, there are no changes in the micro-roughness of the test mirror surfaces as a function of the cleaning process.

In the paragraph given below we provide as additional information the provenience and reference numbers of the Au, Ni, and Rh metal foils as well as the graphite target material used during the e-beam coating process.

- Au foil: Advent Au foil 99.99% purity (reference AU153608).

- Ni foil : Advent Ni foil 99.95% purity (reference NI187908).
- Rh foil: Goodfellow Rh foil 99.9% purity (model LS364067):
- Carbon e-beam deposition target: Goodfellow C sputtering target 99.997% purity (reference C009600).

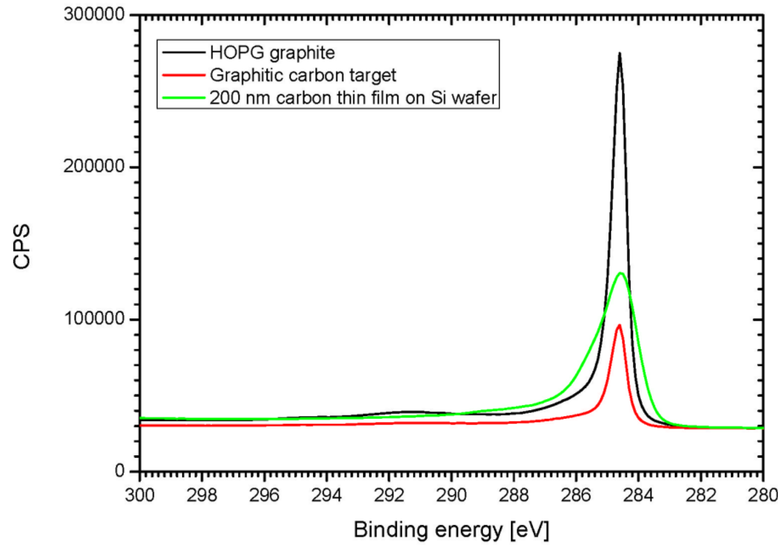

Figure 1 C1s XPS spectra of HOPG, the polycrystalline graphite target material used for the e-beam deposition process, and the resulting carbon thin film as deposed on a Si wafer substrate.

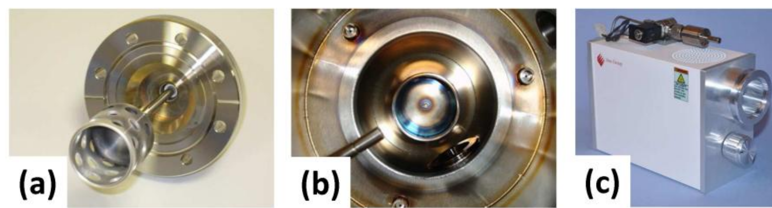

Figure 2 Photographs of the traditional small (a) and large (b) capacitive coupled plasma (CCP) RF guns used for the oxygen plasma cleaning runs. Panel (c) shows the model GV10x inductively coupled plasma (ICP) gun.

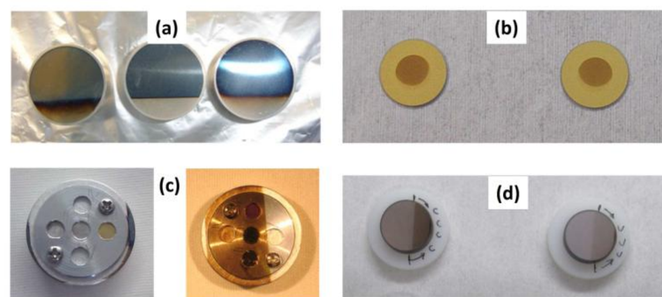

Figure 3 Photographs of: (a) Three aluminium test objects partially coated with a carbon layer (left: 50 nm thickness, centre: 100 nm thickness, right: 200 nm thickness); (b) Two partially carbon-coated quartz micro-balance crystals; (c) An uncoated and a partially carbon-coated holder for metal test foils; (d) Two partially carbon-coated test mirrors (left: Ni reflective coating, right: Rh reflective coating).

Table 1 Micro-roughness data (all in nm rms) from test mirrors with optical quality and different reflective coatings as measured on different spots on the mirror surfaces using interference microscopy before/after an O_2/Ar plasma cleaning. "Rq as made" refers to the micro-roughness figures provided by the manufacturer for the polished Si substrate before reflective coating.

	Mirror #1, Au side			**Mirror #8, Au side**			**Mirror #9, Ni side**		
	Pristine state	After O_2/Ar plasma cleaning		Pristine state	After O_2/Ar plasma cleaning		Pristine state	After O_2/Ar plasma cleaning	
		Carbon coated side	Uncoated side		Carbon coated side	Uncoated side		Carbon coated side	Uncoated side
Rq	0.24	0.19	0.16	0.26	0.18	0.23	0.26	0.23	0.17
Rq	0.28	0.18	0.21	0.27	0.21	0.19	0.25	0.19	0.28
Rq	0.23	0.18	0.20	0.24	0.21	0.16	0.27	0.21	
Rq as made	0.113			0.127			n. a.		

Table 2 Micro-roughness data (all in nm rms) from test mirrors with optical quality and different reflective coatings as measured on different spots on the mirror surfaces using interference microscopy before/after a H_2/Ar plasma cleaning. "Rq as made" refers to the micro-roughness figures provided by the manufacturer for the polished Si substrate before reflective coating.

	Mirror #1, Rh side			**Mirror #8, Au side**			**Mirror #9, Ni side**		
	Pristine state	After H_2/Ar plasma cleaning		Previous state	After H_2/Ar plasma cleaning		Previous state	After H_2/Ar plasma cleaning	
		Carbon coated side	Uncoated side		Carbon coated side	Uncoated side		Carbon coated side	Uncoated side
Rq	0.18 Au	0.169	0.177	0.16 Au	0.160	0.166	0.17 Ni	0.166	0.165
Rq as made	0.113			0.127			n. a.		

[1] J. Diaz, G. Paolicelli, S. Ferrer, and F. Comin, Phys. Rev. B **54** (1996) 8064-8068.
[2] J. E. Blessing, R. E. Ellefson, B. A. Raby, G. A. Brucker, R. K. Waits (2007). J. Vac. Sci. Tecnol. A **25**, 167-186.
[3] SpecLine software, Plasus GmbH, Kissing (Germany).