REMINISCE – Refurbishment of Mirrors to Increase Sustainability at Light Sources

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ABSTRACT

Contamination or damage to X-ray optics is becoming increasingly problematic for beamlines at 3rd and 4th generation storage-rings and FEL sources. Carbon contamination, induced by ultra-intense photon-beams, can cause a major loss in flux, focusing power, and specific absorptions which are detrimental to spectroscopy. For new light sources, with higher coherence, intensity, and repetition rates, contamination and accumulated damage will occur more rapidly. Replacing damaged optics is a time-consuming, risky, and expensive task. Production times are typically >6 months, and sometimes exceed 1 year. Costs per optic are typically tens of thousands of Euro, and can exceed 100k Euro. If several optics need to be replaced on a beamline, the overall cost can be hundreds of thousands of Euros. It is not sustainable or cost effective to replace optics on a yearly basis. Most facilities have independently investigated optical contamination over many years, but coordinated action is now required before the problem gets out of control. These issues motivate two-fold action: find practical methods to slow down the rate of contamination and damage; and develop protocols to safely remove contamination and return the optic to a pristine condition. High-quality metrology and surface science techniques are required to quantify the topography and chemical nature of the contamination, and assess the effectiveness of methods to reduce or remove it. To achieve these challenging goals requires a community effort. We propose an extensive collaboration, including experts from a range of scientific disciplines at various synchrotron and XFEL labs. X-ray optic community projects have recently delivered improvements in the quality X-ray mirrors via enhanced optical metrology. It is hoped that a concerted group effort will deliver similar major advancements, thereby helping to provide beamlines with cleaner, better-performing, X-ray optics.

Keywords: X-ray optics, mirrors, refurbishment, contamination

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1. INTRODUCTION

The reduction of transmission of X-ray optics by irradiation caused carbon contamination is a well-known effect[1] - see also Figure 1 (a SR-mirror after 22 years of use at UE56-SGM-beamline at BESSY-II). With FELs and Synchrotron upgrades to the next generation of diffraction limited storage rings (DLSR), this effect becomes even more significant since the growing rate of such contaminants is proportional to the photon flux[2].[3] Furthermore, such contamination, as well as possible radiation damage, may have an impact on the surface topography and could cause a negative impact on the coherence and could cause scatter. In the past, investigations have also shown subsurface damage below the coating material. Figure 2 shows the subsurface damage on a 510 mm long plane mirror in use for 3 years at the FLASH-FEL at DESY Hamburg. The measurement was taken by means of the BESSY-NOM[4] after the removal of the amorphous carbon coating by cold-plasma treatment. The zone of irradiation damage at the bulk of the mirror substrate can be clearly seen in a 2D slope map.

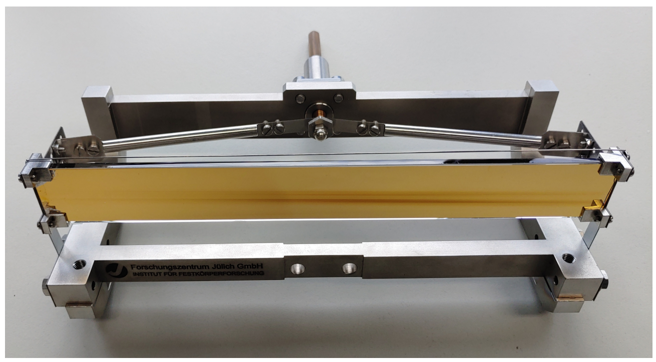

Figure 1. An Au-coated SR-mirror with carbon contamination after 22 years of use at UE56-SGM-beamline at BESSY-II

Investigations to find suitable methods for cleaning optics have been done at most SR- and FEL-facilities, testing various methods. Radio frequency based plasma cleaning (in-situ and ex-situ) is one of the promising methods and UV/ozone based cleaning is another possibility. To extend single tests to systematic studies a sufficient quantity of X-ray optics as sample would be required, which is difficult to realise taking into account the price and delivery times of those optics. For detailed studies of coatings, surfaces and the bulk material before and after the cleaning procedure, very good optical metrology is needed. Topographic and compositional analysis of the surfaces has to be done.

Very promising results have been achieved for smaller optics with plasma-based cleaning[2,5] as well as with UV/ozone cleaning.[6,7] However, those methods have to be adapted and approved for X-ray mirrors with lengths up to one metre as they are used at FELs as well as at storage rings (e.g. as collimating first mirror at bending magnet beamlines at BESSY-II), therefore requiring a homogeneous distribution of the plasma (or ozone) plume.

Furthermore, while studies in the past were focused on classical coating materials in synchrotrons, like gold, platinum or rhodium, new light sources and FELS often use carbon-containing coatings like amorphous-C or boron carbide (B_4C).[8] Only a few studies have been done with carbon based coatings.[9,10]

However, the authors remark that measurements of the surface reflectivity indicate etching effects of the B_4C surface by oxygen plasma. In addition, precaution has to be taken about sputter erosion of nearby surfaces.

Used and carbon-contaminated mirrors, extracted from a beamline after several years of operation only exist in a small amount in each facility. Due to this lack of multiple test specimens in one facility, most studies have been conducted using substrates coated with an amorphous carbon thin film.[11,12] Those studies give a decent insight into the de-contamination process observed. Are these results and methods conferrable to long-term irradiation-based contamination? This has to be investigated. A collaboration of facilities can provide here a collection of contaminated samples and therefore allow for more systematic and large-scale experiments.

Another method of refurbishment is to completely remove the coating layers, without affecting the substrate quality, and subsequently re-apply a fresh coating. This is already a common procedure for mirrors in astronomy, but we lack experience in the X-ray optics field and the right procedure needs to be developed and investigated.

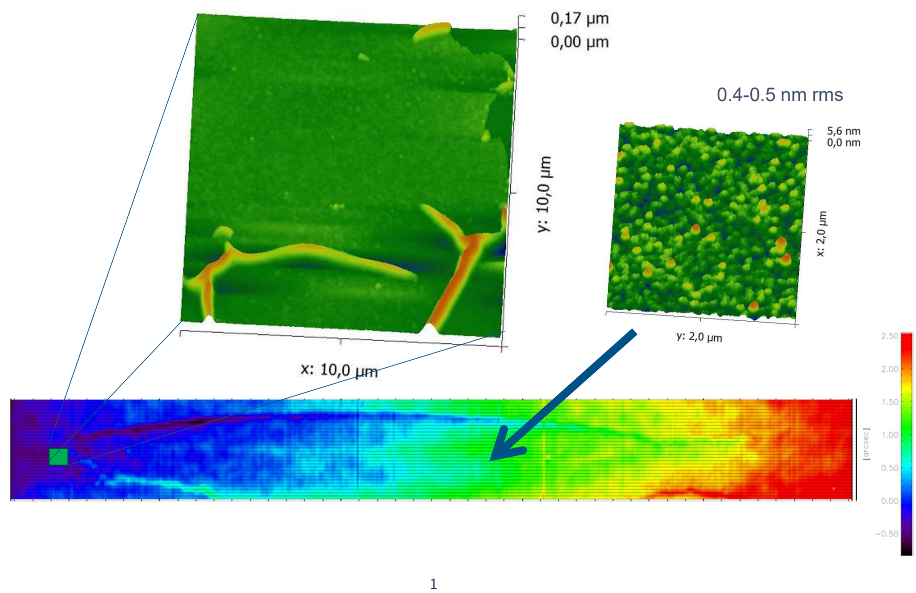

Figure 2. A 510 mm plane mirror after 3 years of use at FLASH. The Fig. shows irradiation damage at the mirror bulk after removal of the a-C-coating by means of cold-oxygen-plasma treatment. The mirror was inspected by slope-mapping at the BESSY-NOM.[4] AFM measurements show significant damage at the edge-section of the irradiated aperture as well as a increase of micro-roughness at the aperture center. Note the micro-roughness at the unirradiated mirror was of 0.2 nm rms.

Moreover, studies of the surface roughness of the underlying substrate material and its sensitivity to the removal process have to be better understood so as not to compromise the polishing quality.

To achieve this understanding and to get the opportunity to enlarge to wide range studies, a collaboration of the synchrotron and FEL metrology community is planned. We plan to join forces among the metrology groups of the light sources, as already done in similar initiatives (MooNpics,[13] LEAPS-INNOV[14]) which successfully led to major achievements and developments in the field of high-precision X-ray mirror fabrication and metrology.

2. FIRST STEPS

The idea of the REMINISCE project was presented during a kick-off meeting on 17 February 2025 in Trieste, taking the opportunity of many interested people from the optics departments being there for the LEAPS INNOV annual meeting. During this meeting, 8 facilities presented their experiences and preliminary studies in cleaning and refurbishment methods of high-precision X-ray optics, and their interests and expectations in this project.

This meeting confirmed that all facilities have started several similar approaches to face this problem. With a collaboration of all facilities, combining the various measurement and laboratory capabilities, these approaches can be extended to systematic, large-scale studies.

As a first step, the project will start with the collection and coordinated dissemination of the existing knowledge in the facilities. Digital solutions like an online platform will be checked for this purpose. Further, several work packages will be created.

- Investigations on and simulation of different types of damage are needed using optical metrology methods. Observations at contaminated mirrors showed differences in the structural appearance of the carbon contamination. The carbon structure will be investigated to understand the chemistry of contamination formation. Main focus will be on contaminated samples collected at the participants' beamlines. But further test samples will be created by short-term X-ray beam exposure. This can be done in the metrology beamlines existing in some facilities like DLS and ESRF. Similar investigations have been done in the EUV Lithography.[15] Other damage tests with higher energies can be performed at the EuXFEL beamlines. Detailed studies will be done on surface physics and chemistry to understand the impact of radiation exposure on the coatings, and the underlying substrate to understand if contamination migrates into the bulk. Several facilities possess instruments like Raman spectroscopy, SEM, EDS, AFM, XRR, and XRD that are suited to do these measurements.

- Development or adaptation of different cleaning procedures, such as plasma technologies, UV cleaning, wet solvent cleaning, and Ion Beam Figuring have to be tested for applicability to the X-ray optics requirements. Test samples will be produced, in part in-house in the facilities that run coating laboratories. The impact of the cleaning process on the high-quality optical surfaces will be determined using the methods mentioned above. Another refurbishment possibility is to remove the coating of a mirror using binding layers that can be etched. Though this method has been used in the past, it is not yet understood if the method also works for optics that have been exposed to X-ray radiation. In preliminary tests at ESRF used mirrors showed a different behaviour than unused ones.

- Prevention of contamination and degradation is an important aspect of the sustainability of X-ray optics. The different possibilities of in-vacuum gas insertion will be investigated.[16] Attention has to be paid to the parasitic deposition of other specimens on the optics' surfaces when using this method. Simple, ideally passive (i.e. without interruption of beam delivery), methods for in-situ monitoring to quantify the amount and type of contamination have to be found.

For all topics, we will further examine solutions that might exist in other areas, like X-ray astronomy or EUV-lithography. The idea is to get in contact with the respective field and adjust the technique for the specific needs of the light sources. Possible industrial partners can be found in the coating industry, producers of plasma chambers or UV/ozone cleaning chambers.

3. PREVIOUS RESULTS

3.1 FERMI

Figure 3: After ten years of operation, the first two plane mirrors of FERMI (PM1a and PM1b), both made of silicon with a 500 Å graphite coating, show clear signs of wear. Plane-wave interferometry (6-inch transmission flat, $\lambda = 632\,\mathrm{nm}$) reveals the presence of surface bumps in the central region of both mirrors. These deformations are more pronounced on PM1a (top), the first optic exposed to the beam, suggesting a key role of thermal load in their development. Follow-up Atomic Force Microscopy revealed an increase in surface roughness from the mirror edges toward the damaged zones. Raman spectroscopy further showed a transition of the carbon in the bump regions towards a more ordered, graphitic-like structure.

3.2 Diamond Light Source

Accurately measuring the location and thickness of contamination on optical surfaces is an essential step to quantify the rate of deposition and the effectiveness of methods to remove it. However, experience has shown that, without due care, metrology data can sometimes give confusing results. When measuring optical surfaces containing more than one type of chemical structure, it is important to understand that the phase unwrappping algorithms used by phase-shifting interferometers will often generate erroneous topographical results.[17] This

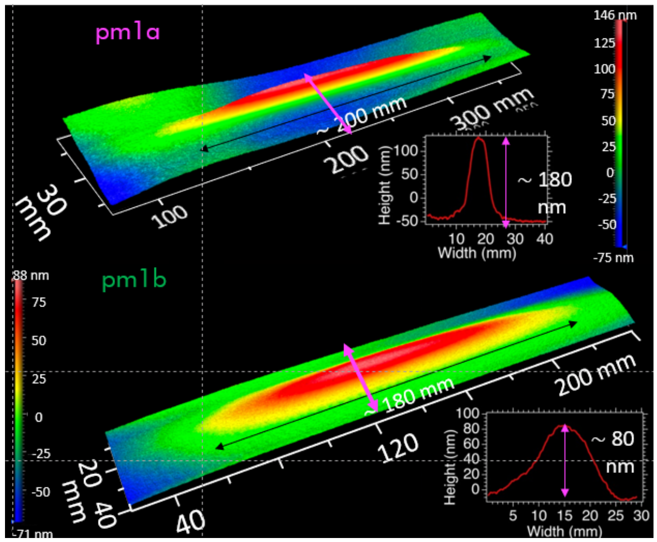

Figure 3. Effects of long-term usage on the first and second plane mirrors of FERMI, revealed by interferometric characterisation

includes optics with several types of coated regions, or carbon contamination covering a portion of the surface. The upper left image in Figure 4 shows the 2D height error of a heavily contaminated X-ray mirror as measured by a Fizeau interferometer. The lower left chart shows a 1D profile along the central tangential line of the mirror. Note that the contamination (blue-coloured regions) appears to be below the plane of the optical surface! However, if a single type of coating with uniform thickness, in this case B_4C, is deposited to cover the entire optical surface, the right image in Figure 4 records a more accurate representation of the surface, including a contamination layer with a thickness of up to $\sim$ 250 nm. The B_4C coating can be readily removed with Cr etchant after testing. Several labs have anecdotally reported that thin layers of contamination, or some forms of defect, are not always visible using interferometry, and only become apparent after final coating.

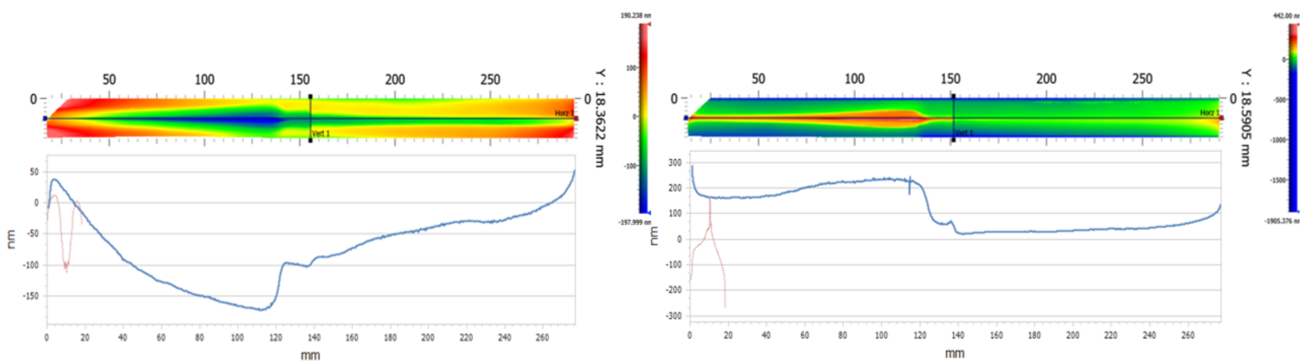

Figure 4. Phase shifting interferometers will often generate erroneous surface profiles when more than one type of chemical structure is present on the optic. The left image falsely indicates that the contaminated region extends into the body of the substrate. However, when a single-type, uniform-thickness, coating is applied to the mirror, the right image reveals a more accurate profile of the (buried) contamination.

Aside from white-light and phase shifting interferometry, scanning probe techniques, such as Atomic Force Microscopy (AFM), can provide useful and complementary information about surface topography of contaminated optical surfaces, especially for length scales <100 microns. Figure 5 shows an AFM scan of a heavily-contaminated monochromator crystal. The right image is a zoomed region of the full scan shown in the left image. AFM reveals important surface structural information about the type and formation of the contamination, especially for features with spatial dimensions below the resolution limit of optical techniques. The "contacting" method of AFM (via van der Waal force interactions) can also reveal the presence of very thin layers of contamination which can be partially, or even fully, transparent to visible light instruments. Such studies emphasise the importance of multi-technique analysis to accurately determine the chemical and structural properties of optical contamination.

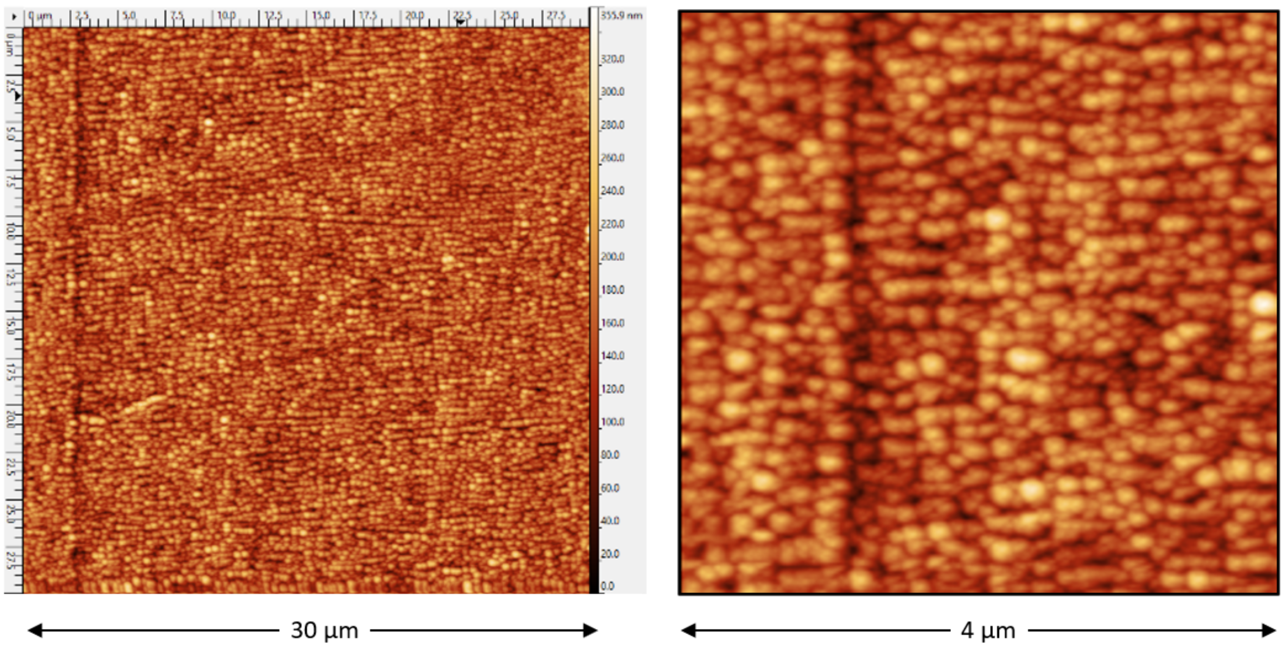

Figure 5. Atomic force microscopy of a monochromator crystal which has been heavily-contaminated by many years of exposure to a polychromatic X-ray beam. The surface topography is dominated by spherical particulates with a diameter between 100 and 300 nm.

3.3 ESRF

The ESRF is a high energy X-ray source with no experimental stations operating with primary beam energy around the C K-edge. As a consequence, the impact on the beam spectrum of the contamination of reflective optics by carbon is limited and typically only affects the transmitted intensity significantly for beamlines exploiting soft X-rays ($E \lesssim 2\,keV$). More problematic is the distortion of the X-ray wavefront due to the presence of inhomogeneous contamination at the optical surface. Such effects are prejudicial to instrument performance both for coherent scattering applications and can also severely degrade the quality of beam focusing even for hard X-rays. The problem is not limited to optics subjected to white-beam illumination but also those exposed only to monochromatic beams. Improvements in the source brilliance following the implementation of the ESRF Extremely Brilliant Source[18] have tended to exacerbate and accelerate these contamination phenomena. The ESRF is exploring various approaches to slowing the degradation through redesign of the optical systems to reduce the exposure of the optical surfaces to carbon-rich contaminants. In parallel, efforts have been made to restore the quality of the optics using ex-situ cleaning, coating-stripping and re-coating techniques. For ex-situ cleaning, trials using oxygen plasma, UV-Ozone, carbon dioxide snow and polymer film cleaning methods have been performed on both both bare silicon and functionally coated surfaces (with total-reflection and multilayer coatings). An important consideration in the application of any of these methods is that they should minimally impact the underlying quality of the pristine optic. It is particularly important not to degrade the roughness of the reflecting surface which may be $\sim 0.1\ nm$ rms. Thus far, the trials have indicated that several techniques satisfy this requirement and appear capable of reducing or eliminating the contaminating layer thickness, but no 'universal' recipe has been found to be suitable to restore the quality of all contaminated optics. Figure 6 shows a heavily degraded single crystal silicon monochromator optic showing the effectiveness of UV-Ozone cleaning in progressively reducing the visible contamination trace. In this instance the crystal quality was completely recovered. Apparently similar contamination on uncoated mirror optics does not respond to ozone exposure in the same manner. A better understanding of the nature of the contamination is clearly required. Figure 7 illustrates the behaviour of a nickel coated focusing mirror following treatment with oxygen plasma. Significant reduction in the 'stripe' contamination is observed but faint traces persist which are not removed even after prolonged exposure to the cleaning process.

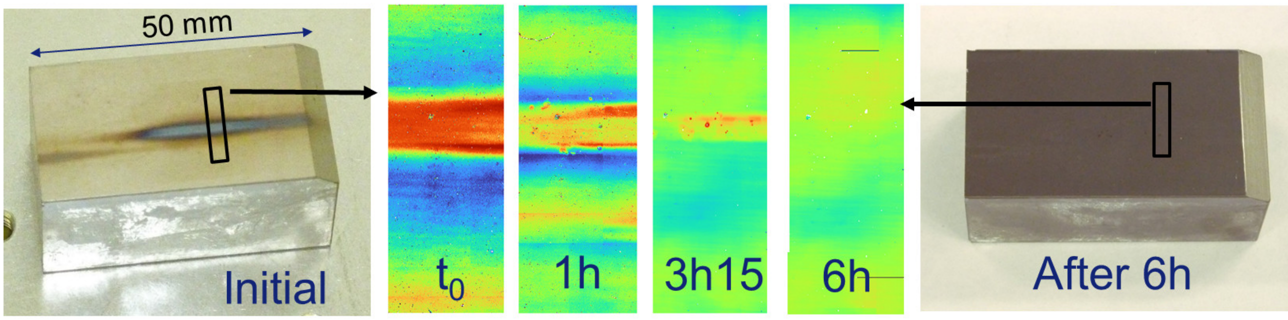

Figure 6. UV Ozone Cleaning

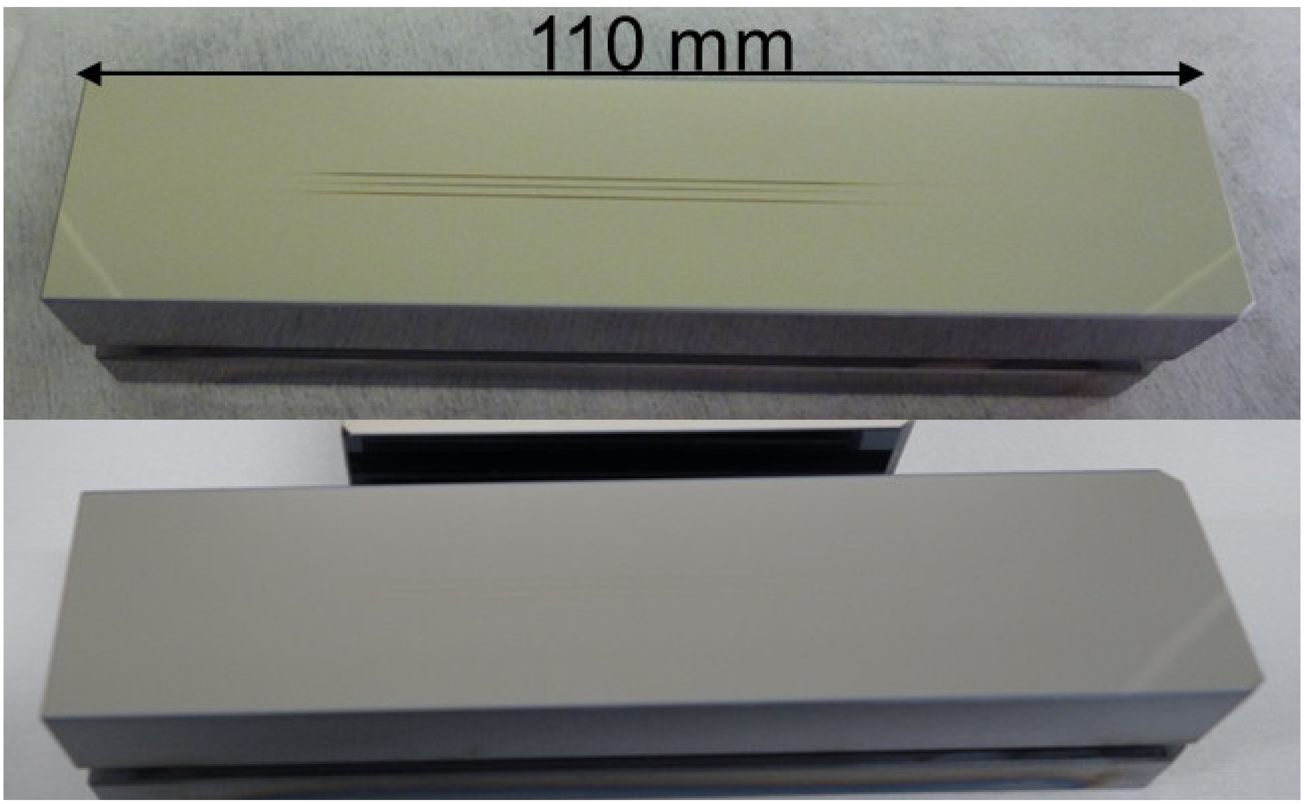

Figure 7. Oxygen Plasma Cleaning

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