

UV PERFORMANCES OF PULSED LASER DEPOSITION GROWN Mg PHOTOCATHODES*

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Abstract

We report a detailed description of the laser cleaning procedure and emission performance measurement on a Pulsed Laser Deposited Mg film. During the tests performed after the end of each cleaning operation we have evidenced an increase of Quantum Efficiency (QE) in time. Then the QE apparently stabilizes at a remarkably higher value. The study of this phenomenon is important because it determines both the working QE value and the lifetime of the cathode. Moreover, the stability of the QE has been revealed for a time scale of several days after each laser cleaning process, in our vacuum conditions.

INTRODUCTION

The electron injectors for the advanced projects of fourth generation X-ray sources, as SASE-FELs [1], and for future linear colliders [2], are mostly based on laser excited photocathodes. Metals have order of femto-second response and they are rugged in handling. Among them, Mg has premium QE, in the order of 10^{-3} at near UV wavelength (266 nm). However, bulk Mg cathodes have shown poor emission uniformity and failures in the high electric fields environment of RF guns. Moreover, the QE value degrades in time even in the UHV vacuum of RF guns. As an alternative, Mg films on copper substrates have been proposed and tested on the basis of their presumed better purity and uniformity [3]. Previous reports on Mg films grown by Pulsed Laser Deposition showed promising results in particular with respect to uniformity of emission, quantum yield, and adhesion to the substrate [4-6].

In order to reach the utmost QE value is necessary to remove the inevitable oxide layer that forms on the film surface. Only two techniques were used to perform such operation: laser cleaning and ionic etching [7]. The laser cleaning operation must be performed with particular care due to the limited thickness of the film and to avoid inducing surface roughness. It was evidenced that the cleaning irradiation may determine a variation of the film surface morphology [8].

The aim of this paper is to describe the film deposition conditions, the laser cleaning procedure and to evidence the performance of Mg films from photoemission point of view.

EXPERIMENTAL

Cathode preparation

The PLD technique is a well-known method for thin

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films deposition of several materials [9, 10]. The PLD apparatus used to deposit thin films is shown in figure 1. During the deposition the vacuum chamber was filled with pure He at a pressure of 5 Pa to reach the plume confinement regime leading to an increase of the deposition rate [11].

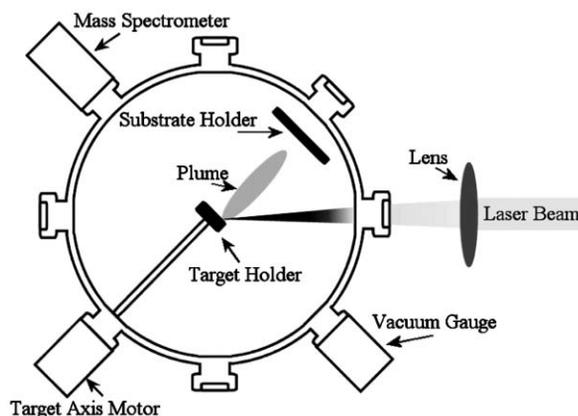


Figure 1: PLD experimental setup.

With the aim to clean the target surface, a pre-irradiation treatment was applied with 5,000 laser pulses. During this laser cleaning, the Cu substrate was shielded from the initial ablated material, which could contain impurities. Several films were prepared to optimize the deposition process. The deposition parameters of the tested sample are shown in table 1.

Table 1: Mg film deposition parameters.

Target	Mg
Substrate	Cu
Target-Substrate distance	3.5 cm
Laser spot size	1.0 mm ²
Base pressure	5 x 10 ⁻⁶ Pa
Laser pulses	
• Cleaning (5 x 10 ⁻⁶ Pa)	5000
• Deposition (in He at 5 Pa)	50000
Laser fluence	10 J/cm ²
Film thickness	2.5 μm
Film diameter	12 mm

Vacuum diode cell

The QE measurements were performed in a photodiode cell under UHV condition. The cathode and the anode separated at a distance of 3 mm were placed inside the photodiode cell (fig. 2). The Mg film occupied the cathode position and was electrically grounded. The anode plate was biased with high DC voltages up to 5 kV thus allowing the generation inside the gap of an intense electric field of about 1.7 MVolt/m.

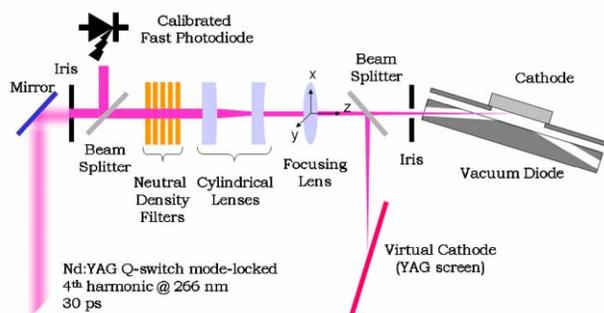


Figure 2: Laser cleaning and QE measurement setup.

In order to illuminate the cathode and for precise alignment of UV laser beam on cathode surface the anode plate was machined with two symmetric holes. They have a diameter of 4 mm and form an angle of 72° with respect to the normal of the cathode surface. This particular geometry was chosen for two main reasons:

- The first was to avoid the use of a metallic grid usually present in such experiments. Our previous experimental results clearly indicated that the light distribution on the cathode surface, in presence of a metallic grid, was affected by diffraction structures and hampered uniform illumination of the cathode surface [6];
- Secondly, the angle of 72° was chosen in order to perform experiments with the same geometrical configuration used in the last generation of the BNL/SLAC/UCLA 1.6 cells S-Band RF gun [12].

The vacuum chamber in which the photodiode cell was inserted was evacuated at a base pressure of about 2×10^{-7} Pa by means of an ionic pump. The quality of the vacuum was controlled by a quadrupole mass spectrometer.

Laser cleaning and QE measurements

Laser cleaning and QE measurements of the Mg films were performed using the UV radiation of the 4th harmonic of a Q-switch, mode-locked Nd:YAG laser (QUANTEL YG-501) able to deliver up to 300 μ Joule at 266 nm with a pulse duration of 30 ps. The laser energy measurements were performed by integrating the signal of the calibrated fast photodiode, while the charge measurements were done using a charge integrator.

The laser cleaning procedure was performed by scanning the focused laser beam over an area of about 2.4×2.4 mm². The laser beam had a diameter on the cathode surface of about 300 μ m and energy of about 25 μ Joule per pulse (power density and laser fluence were about 3 GW/cm² and 35 mJ/cm² per pulse respectively). Such value of the power density was experimentally determined during preliminary tests as being the threshold for the ablation of the oxidized layers. The power density threshold value for the ablation was deduced looking at the vacuum level of the UHV chamber: we observed that once such value was reached, the vacuum level inside the chamber slightly worsened indicating that material ablation was occurring.

FEL Technology I

The scanning scheme was chosen in such way that laser shots focused over consecutive horizontal path lines slightly overlap in order to make sure that all parts of the surface were exposed to the laser beam. Typically during the laser cleaning the laser repetition rate was set at 5 Hz and the scanning speed of the laser spot was on the order of 10^{-3} m/s.

After each laser cleaning procedure the focusing lens was removed and the laser beam was directed to the central part of the irradiated area of the cathode to measure the QE. In order to perform measurements far from space charge saturation, the laser energy was decreased by means of calibrated neutral density filters.

RESULTS

The first emission measurement was performed before any laser cleaning treatment. It revealed a very poor QE of about 2×10^{-6} . Just after the first laser cleaning procedure, performed as above described, the QE measured value rose of about 2 orders of magnitude reaching the noticeable value of 5×10^{-4} , very close to the value reported in literature for bulk pure Mg [13].

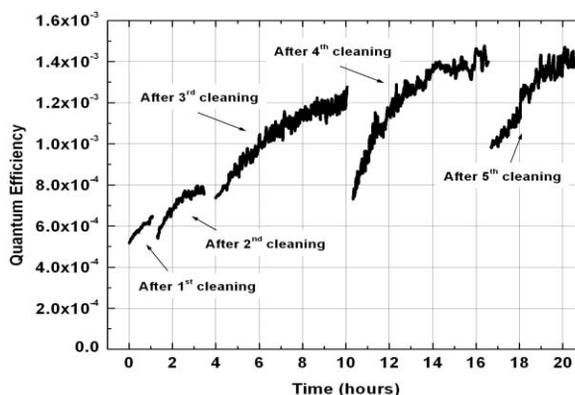


Figure 3: QE measurement vs. time after laser cleaning.

In order to study the stability of emission we performed QE measurements vs. time for several hours (fig. 3). The measurement after the first laser cleaning was performed for about 1.5 hours. We detected a slight increase of the QE value during this time from the initial value (just after the cleaning) of about 5×10^{-4} up to 6.5×10^{-4} . After about 12 hours, we carried out a new laser cleaning of the cathode surface. The QE value was monitored again for about 2.5 hours. After the 2nd cleaning, the measured QE was slightly lower than the last measured value, but also in this case its value increased from 5.5×10^{-4} of up to about 8.0×10^{-4} . About 12 hours later, we executed a 3rd cleaning and measured the QE continuously. We observed again that after an initial decrease just after the laser cleaning (from 8.0×10^{-4} to 7.5×10^{-4}), the QE value increased up to 1.2×10^{-3} during the next 6 hours. The 4th cleaning procedure was accomplished about 12 hours before the last QE measurements. In this case, the single raster scheme was performed 5 times. After the laser cleaning we detected a strong decrease of the QE that lowered to an initial value

of 7.5×10^{-4} . During the next 6.5 hours we continuously measured the QE value, observing a continuous increase up to a value of about 1.4×10^{-3} . The 5th and final laser cleaning, carried out using a single raster scheme, resulted in an initial QE value of 10^{-3} that rose up to 1.4×10^{-3} during next 4 hours. It is evident the strong variation, up to a factor two, during each measurement.

The sample was kept under vacuum at a base pressure of 2×10^{-7} Pa during the next two days. After this time we performed new measurements without additional laser cleaning procedure. We observed that the QE value was the same with that measured two days before, i.e. 1.4×10^{-3} . This value was preserved also after the irradiation of the surface with about 5 μ Joule for about 1 hour with a repetition rate of 5 Hz. The aim of the test was to verify the ability of such cathode to maintain its emission performances also during irradiation with laser energy slightly exceeding that required to extract electron bunches of 1 nC charge.

Additionally the QE map of the irradiated area was drawn. In order to perform such measurement the laser energy was lowered to few nJ by means of neutral density filters. The laser was focused on the cathode surface by a 30 cm focal length fused silica lens. The truncated gaussian laser spot was then moved in succession over a grid of 9x9 points on the laser-cleaned area. The average QE on each irradiated area of about 300 μ m diameter is evaluated from the slope of emission curves, i.e. collected charge vs. laser energy, far from space charge saturation regime. The 300 μ m diameter is the minimum compatible with the sensitivity of our charge detector apparatus to remain below the extractable charge limit. Naturally this entails a limit on the spatial resolution which cannot be less than about twice the value of the laser spot diameter.

The QE map of the sample collected 72 hours after the 5th and last laser cleaning process is reported in figure 4. With the limitations due to the size of the laser spot during such measurements the emission seems to be reasonably uniform inside a circle of about 500 μ m of radius centred in the most intense emitting area.

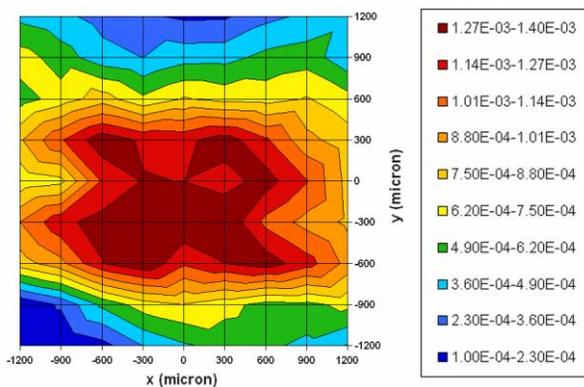


Figure 4: QE map after 5th laser cleaning.

DISCUSSION

QE values for Mg reported by now in literature, either for bulk or film, are scattered in a range within 2.5×10^{-4}

and 3×10^{-3} for an exciting wavelength of 266 nm. The reason of this spread can be the different experimental conditions, as e.g. vacuum base pressure and electric field intensities [14-16].

Theoretical prediction for the QE value for Mg at 266 nm indicates a value of about 5×10^{-4} . Higher values cannot be predicted assuming a work function of 3.66 eV. A decrease of such value by means of the formation of some electric dipole on the surface due to the gas absorption may be the key to explain such high photoemission performance from this material.

The very gradual laser cleaning procedure described before should remove only the uppermost deeply oxidized layer. It has the typical rock-salt structure of the MgO and was formed by exposing the film surface to air during the time between the deposition procedure and emission characterization, both performed in UHV environment. Ref. 3 reports that successive laser cleaning procedures resulted in an increase of the QE. This is also clearly observed also in our previous [4-6] and present experiments, demonstrating that pure Mg metal surface has been obtained with the removal of surface oxidized layers. Nevertheless, a continuous monitoring of the emission yield of the Mg film during a time interval of several hours after each laser cleaning clearly indicates that some other processes influenced the electron surface emission properties. Few years ago the effects of oxygen adsorption on the quantum yield of Mg films have been reported by Q. Yuan et al. [17]. They showed that a significant increase of QE is achieved by exposing Mg film to controlled fluxes of pure O₂. After an initial increase, strongly dependent on the oxygen dose, the quantum yield deteriorates, pointing to the formation of a thick oxidized layer that delayed the emission of photoelectrons. Similar results were observed in 1935 by R.J. Cashman and W.S. Huxford [18]. Their findings on the lowering of the work function during initial oxidation of the Mg surface were confirmed through numerical computation by E. Schröder et al. [19].

Our experimental situation was quite different from those of the experiments reported in literature. In our case no fluxes of oxygen have been introduced in the vacuum chamber. Our working base pressure was 2.5×10^{-7} Pa and the O₂ partial pressure was two orders of magnitude lower than that used in other experiments reported in literature [17, 18]. Partial pressures of residual gases were measured by a quadrupole mass spectrometer. From the residual gas mass spectra we observed that the most abundant chemical species were H₂, H₂O, N₂ and O₂, with partial pressures of 1.9×10^{-7} Pa, 1.5×10^{-8} Pa, 3×10^{-8} Pa and 8×10^{-9} Pa, respectively.

According to our readings of partial pressure and to the duration of the QE acquisitions, which, as reported in figure 5, were performed up to a maximum of about 6 hours, we deduced that the Mg surface exposed to O₂, at partial pressure level of 8×10^{-9} Pa, led to a surface coverage lower than 1 monolayer. On the contrary, H₂ exposure during the same time, due to higher partial pressure of this chemical species with respect to

molecular oxygen, may give rise to a surface coverage higher than 2-3 monolayers. According to R. J. Cashman and W. S. Huxford [18] who studied the emission properties of Mg surface exposed to H₂ fluxes, we consider that the increase of the quantum yield of our sample is related to the chemisorption of hydrogen in subsurface sites of Mg film, and in a lesser extent of oxygen. This should lead to a decrease of the work function because an electric dipole is formed under those conditions near the surface of the film with orientation that helps photoexcited electrons to escape in vacuum. Such idea is partially confirmed by the findings of P. T. Sprunger and E. W. Plummer [20] who observed a decrease of 0.95 eV of the initial value of the work function after the complete hydrogenation of the Mg(0001) surface due to the formation of a metastable "hydride" phase. They also reported that the activation energy for the chemisorption of H₂ on Mg(0001) surface was of about 0.5 eV and thus at room temperature the formation of this phase should not be observed. PLD films grown at room temperature usually do not present a well defined crystalline structure. XRD spectra (here not showed) reveal that polycrystalline Mg film was formed on laser cleaned areas with slight texture on (002) orientation. Under these conditions, we can expect that the activation energy for chemisorption of H₂ on Mg amorphous surfaces may be lower than that reported for Mg(0001), as also indicated by Nørskov et al. [21].

Tests performed by irradiating the cleaned area with laser pulses ($\lambda=266$ nm, $\tau=30$ ps) having 5 μ J of energy show any influence on the QE. This result seems to indicate that the energy required to photo-dissociate the Mg-H bond should be higher than 4.67 eV.

The emission inside the central part of the laser cleaned area within a radius of approximately 500 μ m and considering the limitations of our experimental arrangements seems to be uniform within 20%. We found out that the rapid decrease of the efficiency observed for vertical coordinate values close to the extreme of the measured area is just due to a non-perfect alignment of the laser beam inside the *bore* where it has to travel to illuminate the cathode surface. This misalignment does not influence the measurements in the central zone of the scanned area.



Figure 5: Mg film deposited over the backflange of a BNL/SLAC/UCLA 1.6 cells S-Band RF gun.

CONCLUSIONS

We presented the results of laser cleaning effect in terms of Quantum Efficiency, emission uniformity, surface morphology and structure of pulsed laser deposited pure Mg films on copper substrate. Moreover, the influence of the chemisorption of hydrogen, always present as residual gas in ultra high vacuum operating conditions, has been discussed with particular attention with respect to the efficiency and stability of the emission performances of Mg surface. Mg films have been grown in the central part of a backflange suitable to be inserted onto an RF gun and the power tests are in progress (figure 5). This work has been partially supported by the EU commission in the sixth framework program, contract no. 011935 EUROFEL and by MIUR, Progetti Strategici, DD 1834, December 4, 2002.

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