Available online at www.sciencedirect.com
SCIENCE DIRECT®
Applied Surface Science 248 (2005) 397–401
applied surface science
www.elsevier.com/locate/apsusc

Pulsed laser deposition of Mg thin films on Cu substrates for photocathode applications

L. Cultrera [a,*], A. Pereira [b], C. Ristoscu [c], A. Clozza [d], F. Tazzioli [d], C. Vicario [d]

[a] University of Lecce, Physics Department and INFN, Via Arnesano, 73100-Lecce, Italy
[b] LP3, CNRS/University of Mediterranee, 163 Avenue de Luminy C917, 13288 Marseille, France
[c] Lasers Department, National Institute for Lasers, Plasma and Radiation Physics, P.O. Box MG-36, Bucharest V RO-77125, Romania
[d] Istituto Nazionale di Fisica Nucleare (INFN), Laboratori Nazionali di Frascati, Via E. Fermi, 40-00044 Frascati, Italy

Available online 11 April 2005

Abstract

In this study, pulsed laser ablation of pure Mg targets (99.99%) using an XeCl excimer laser ($\lambda = 308$ nm, $\tau = 30$ ns) was used to grow Mg coatings on Cu substrates. A thin layer of Mg (100–200 nm) was deposited in UHV of 5×10^{-6} Pa to ensure high purity. A second ultra-thin layer of MgO of 10–20 nm was deposited in oxygen at 10^{-2} Pa pressure to protect the previous layer from atmospheric contamination.

Measurements for determining the electron yield of the deposited samples under UV irradiation were performed on a diode structure in another UHV apparatus evacuated to 10^{-6} Pa.

Keywords: Laser deposition; Photocathode applications; Mg thin films

1. Introduction

High energy physics accelerators and free electron lasers put increased demands on electron beam sources. Cesium telluride (Cs_2Te) photocathodes can operate for 3–6 months with a quantum efficiency (QE) higher than 7% and for 2 years with a QE higher than 2%, in typical RF-guns at a vacuum of 10^{-8} Pa and accelerating fields of 100 MV/m. These cathodes, however, have a shortcoming, i.e., they do not survive for even very short exposure times in vacuum of 10^{-5} Pa [1].

Metal photocathodes are good candidates for generating short-pulse, high-current-density electron beams due to the large free-electron density and their instantaneous response to irradiation. Metallic photocathodes have QE higher than 5×10^{-4} and Mg has also a relatively high UV damage threshold, ~200 μJ/mm^2 to Nd:YAG, $\lambda = 266$ nm, $\tau = 10$ ps [2,3]. These features make Mg a good candidate for photocathode applications. Using of bulk Mg in RF-guns is limited, however, because the Mg-cathode and Cu-support

* Corresponding author. Tel.: +39 0832 297470; fax: +39 0832 297505.
E-mail address: luca.cultrera@le.infn.it (L. Cultrera).

0169-4332/$ – see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.apsusc.2005.03.065

plate junction break often [4]. Thus, using Mg coated Cu cathodes seems to be promising by providing enhanced adhesion of the photo-emissive layer to the substrates.

Still, the strong reactivity of the Mg to oxygen may lead to the formation of oxidized layers reducing the QE of the metallic layer. The preparation of these cathodes requires UHV deposition techniques, thus the pulsed laser ablation and deposition (PLAD) is a powerful tool to synthesize high purity coatings. This method has been already used successfully for depositing various materials such as semiconductors, oxides, nitrides, carbides, alloys and ceramics [5]. In this study, we report the growth of Mg–MgO coatings on Cu substrates by PLAD, for photocathode applications. To our best knowledge, this is the first work reporting on Mg coatings by PLAD.

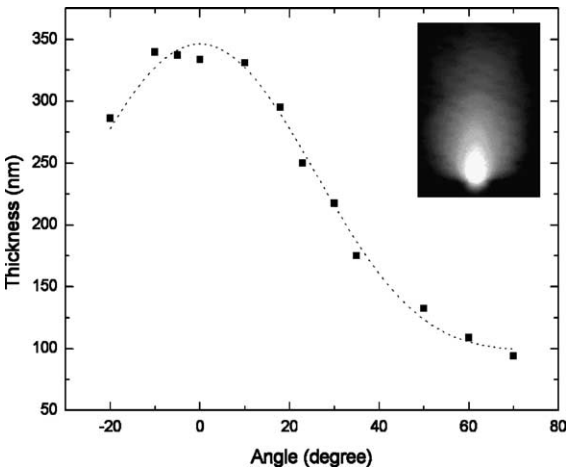

Fig. 1. Mg film thickness distribution as a function of the angle with respect to the normal of the target surface, using an hemicylindrical substrate holder, at the fluence of 9 J/cm^2 and the target–substrate distance of 4 cm. The inset shows a picture of the expanding plume.

2. Samples preparation

PLAD experiments were carried out in an UHV vacuum chamber pumped to a base pressure of 5.0×10^{-6} Pa using a turbomolecular pump. The quality of the residual gas in the chamber was controlled by a quadrupole mass spectrometer (Ametek MA 100). The laser source used was a Lambda Physics LPX 315i XeCl excimer laser operating at 308 nm with pulse duration of 30 ns and repetition rate of 10 Hz.

The Mg targets of 99.99% purity were mounted on a rotating holder to avoid the formation of craters. The ablation process creates a circular crater on the target surface. The focal spot on the Mg target had an area of ~1.2 mm^2 and the laser fluences were 5 and 9 J/cm^2. The plume formed during the ablation experiments was clearly visible and recorded from the top flange of the vacuum system by a digital camera (Panasonic NV-DS28EG) to investigate the plume expansion details. No deflection was detected in the symmetry of the luminous distribution of the ablated material and the material deposited using an hemicylindrical target holder was distributed along the target normal, see Fig. 1 [6,7]. Thus, the conventional configuration, i.e., parallel substrate facing the target's surface, was used in the deposition experiments.

To avoid contamination of the deposited films by the atmospheric gas compounds, O_2, N_2, H_2O, and hydrocarbons, a protective coating of MgO was deposited by ablating the Mg target in high purity oxygen (99.99%). The experimental conditions used for the depositions are summarized in Table 1.

The cleaning of the target surface was performed with $N = 2000$ laser pulses which removed the contaminated surface layers. During this step, the

Table 1
Experimental parameters used for the deposition of samples A and B

	Sample A	Sample B
Target	Mg	Mg
Substrate	Cu	Cu
Target–substrate distance (cm)	6	6
Base pressure (Pa)	5.0×10^{-6}	5.0×10^{-6}
Laser energy per pulse (mJ)	60	110
Laser spot size (mm^2)	1.2	1.2
Laser fluence (J/cm^2)	5.0	9.0
Laser pulses number, N		
Cleaning Mg target (5×10^{-6} Pa)	2000	2000
Deposition of Mg layer (5×10^{-6} Pa)	10000	10000
Covering by MgO layer (O_2 10^{-2} Pa)	2000	2000
Mg layer thickness (nm)	100	200
MgO layer thickness (nm)	10	20

Cu substrate was shadowed by a mask to avoid deposition. The Mg layers were deposited by $N = 10^4$ laser pulses. This gave a thickness of 100 and 200 nm for the sample deposited at laser fluences of 5 and 9 J/cm^2, respectively. Both the cleaning and the deposition process were carried out in 5×10^{-6} Pa UHV. A second thin layer of 10 and 20 nm for laser fluences of 5 and 9 J/cm^2, respectively, was deposited in low oxygen pressure (10^{-2} Pa), creating a thin oxidized layer to protect the previous layer from atmospheric contamination. Previous studies on the dynamics of oxidation of pure Mg showed that the growth of the oxide layer is very fast initially, but proceeds very slowly after that a few nm of Mg are oxidised [8].

The strong reactivity of Mg to oxygen was revealed by a quadrupole mass spectrometer. The intensity of the peaks relative to H_2O and O_2 strongly decreased during the cleaning procedure, while the intensity of the peak relative to N_2 remained constant. Before the depositions, the partial pressures of O_2 and H_2O were 1×10^{-7} and 3×10^{-7} Pa, respectively, while after the depositions their values were 1×10^{-8} and 3×10^{-8} Pa. This indicates that the oxygen reacts with the ablated and deposited Mg in the chamber.

3. Samples characterization

The samples were tested as photocathodes with a dc field of 1 MV/m after cleaning the oxidized layer with high energy laser pulses produced by a mode locked Nd:YAG laser ($\lambda = 266$ nm, $\tau = 30$ ps) in an UHV apparatus evacuated to a base pressure of 10^{-6} Pa.

The surface of sample A was cleaned to remove the oxidized layer with $N = 1000$ laser pulses, each with 100 μJ energy and with a spot size of 0.7 mm diameter; the resulting power density was ∼8 GW/cm^2 (laser fluence, 1.4 mJ/cm^2). The beam was scanned over a circular area having a diameter of 2 mm. The emitted charge as a function of the laser energy before and after the cleaning procedure is shown in Fig. 2. The QE achieved was 1.5×10^{-5}. After further $N = 4000$ cleaning pulses a permanent decrease of the photoelectron emission was detected. Visual inspection revealed that the surface of the Mg coating was damaged in this case. Scanning electron microscope (SEM) characterization of the irradiated surface of sample A showed that the Mg layer was completely removed leaving the Cu surface exposed to the laser beam, see Fig. 3.

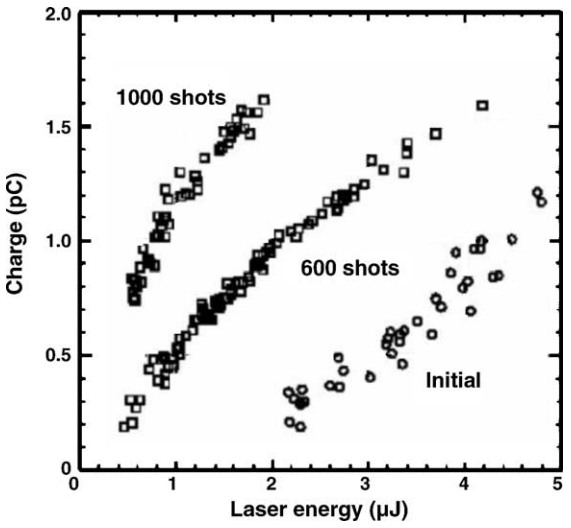

Fig. 2. Emitted charge as a function of laser energy for sample A. Three series of data are shown: before cleaning the oxidized layer, and after removing the oxidized layer with $N = 600$ and 1000 laser pulses.

The photoemission of sample B was studied before and after cleaning with $N = 3000$ laser pulses, see Fig. 4. No perforation of this coating occurred during laser irradiation as confirmed by SEM observations. However, the QE measured for this sample was 1.5×10^{-5}, comparable with that of sample A.

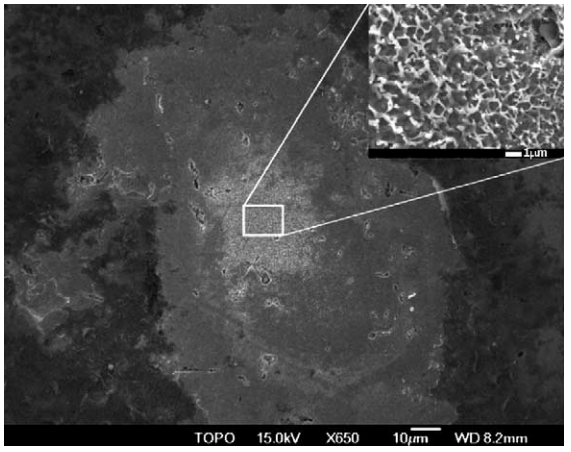

Fig. 3. SEM picture of the irradiated area of sample A. The cleaning procedure completely removed the Mg layer leaving the Cu substrate surface exposed to the laser beam. The inset shows the Cu surface melted by the action of the laser beam.

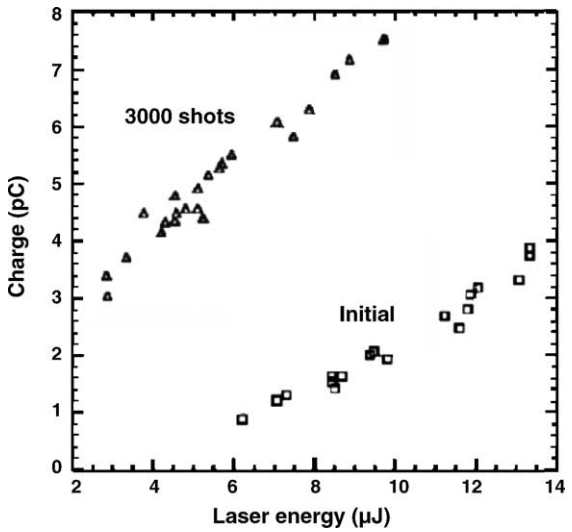

Fig. 4. Emitted charge as a function of laser energy for sample B, before cleaning the oxidized layer and after removing the oxidized layer with $N = 3000$ laser pulses.

4. Discussion

The Mg coatings deposited with PLAD have good potential for photocathode emitters. The absence of plume deflection and the symmetry of the mass distribution of the ablated materials enabled us to deposit Mg coatings by the conventional deposition configuration, with a deposition rate of 0.2 Å per pulse, as calculated from the thickness measurements, using a fluence of 9 J/cm^2 and target to substrate distance of 6 cm.

The reaction of the oxygen-containing chemical species present in the background gas with the Mg removed during the cleaning procedure was helpful in reducing the partial pressure of oxygen-containing species (mainly O_2 and H_2O), which react with the Mg during the deposition process on the Cu substrate. Maximum values for QE are about one order of magnitude lower than those reported in literature for pure Mg metal, e.g., 6.2×10^{-4} [9], indicating that our deposited films consist of a mixture of Mg and MgO. This is in agreement with the Auger analysis reported in the literature for commercially sputtered Mg cathodes, indicating that in a 20 μm thick sample the oxygen contamination is present to a depth of 150 nm under the surface [3], but in contrast with studies on Mg oxidation dynamics [8], which predict only a few nm of oxidized layer. Thus, we conclude that MgO inclusions are in the film due to contamination during the sample preparation. The importance of high vacuum in the preparation conditions of the layers is crucial. In fact, the strong reactivity of the ablated material during the target cleaning procedure with oxygen-containing chemical species was revealed by mass spectrometric studies.

SEM observations confirm that the Mg layer can be removed during the cleaning procedure for QE measurements, leaving the substrate surface exposed to the laser beam (Fig. 3). Thicker films can avoid removing of the whole Mg film when cleaning the oxidised layer.

We conclude, thus, that although the deposition experiments were performed in an UHV apparatus, the Mg thin film growth on the Cu substrate reacted with oxygen thus including also MgO. The purity of the films and, hence, their performances as photocathodes can be strongly improved by lowering the base pressure to 10^{-7} Pa level.

5. Conclusions

PLAD of Mg coatings on Cu substrate gave promising results. Improvements could be obtained by protecting the Mg surface with a covering layer, such as graphite, that could be carried out in UHV conditions without an increase of the pressure inside the chamber, which occurs in the case of MgO deposition.

Acknowledgement

The authors wish to acknowledge the support to this research given by Istituto Nazionale di Fisica Nucleare (INFN).

References

[1] I. Boscolo, P. Michelato, Nucl. Instrum. Methods Phys. Res. A 445 (2000) 389.
[2] T. Srinivasan-Rao, J. Fischer, T. Tsang, J. Appl. Phys. 77 (1995) 1275.

[3] T. Srinivasan-Rao, J. Schill, I. Ben Zvi, M. Woodle, Rev. Sci. Instrum. 69 (1998) 2292.
[4] T. Srinivasan-Rao, D.T. Palmer, I. Ben-Zvi, R.H. Miller, X.J. Wang, M. Woodle, Presented at the Particle Accelerator Conference, Vancouver, Canada, 12–16 May, 1997 (Paper 3W–20).
[5] D.B. Chrisey, G.K. Hubler, Pulsed Laser Deposition of Thin Films, Wiley, New York, 1994.
[6] A. Perrone, A. Zocco, L. Cultrera, D. Guido, A. Forleo, Appl. Surf. Sci. 197–198 (2002) 251.
[7] L. Cultrera, D. Guido, A. Perrone, M.I. Zeifman, Appl. Phys. A 79 (2004) 1181.
[8] T. Do, S.J. Splinter, C. Shen, N.S. McIntyre, Surf. Sci. 387 (1997) 192.
[9] T. Srinivasan-Rao, J. Fischer, T. Tsang, J. Appl. Phys. 69 (1991) 3291.