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Tailoring the emissive properties of photocathodes through materials engineering: Ultra-thin multilayers

Daniel Velázquez [a], Rachel Seibert [a], Hasitha Ganegoda [a], Daniel Olive [a,b,c], Amy Rice [a], Kevin Logan [a], Zikri Yusof [a], Linda Spentzouris [a], Jeff Terry [a,*]

[a] Physics Department, Illinois Institute of Technology, Chicago, IL 60616, USA
[b] Chemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA
[c] Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

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ABSTRACT

We report on an experimental verification that emission properties of photocathodes can be manipulated through the engineering of the surface electronic structure. Ultrathin multilayered MgO/Ag(0 0 1)/MgO films were grown by pulsed laser deposition, tuning the thickness n of the flanking MgO layers to 0, 2, 3, and 4 monolayers. We observed an increase in quantum efficiency and simultaneous decrease in work function with layer thickness. The scale and trend direction of measurements are in good but not excellent agreement with theory. Angle resolved photoemission data for the multilayered sample $n = 3$ showed that the emission profile has a metallic-like momentum dispersion. Deviations from theoretical predictions [K. Németh et al., PRL 104, 046801 (2010)] are attributed to imperfections of real surfaces in contrast with the ideal surfaces of the calculation. Photoemissive properties of cathodes are critical for electron beam applications such as photoinjectors for Free Electron Lasers (FEL) and Energy Recovery Linacs (ERL). An ideal photoemitter has a high quantum efficiency, low work function, low intrinsic emittance and long lifetime. It has been demonstrated here that emission properties may be systematically tailored by control of layer thickness in ultrathin multilayered structures. The reproducibility of the emission parameters under specific growth conditions is excellent, even though the interfaces themselves have varying degrees of roughness.

1. Introduction

Much of the development of photocathode materials has been aimed at the growth of photoemissive thin films with low work function (WF), and high quantum efficiency (QE) [1]. It has been shown, both theoretically and experimentally, that metal-insulator junctions can lead to the modification of the WF for coverages of a few monolayers (ML) of metal oxides on metallic substrates [2–6]. Reduction of WF and increase of QE can be achieved simultaneously by coating metal surfaces with Cs, CsBr, Y, and Mg[7–11]. Cs ion implantation on Cu, Ag, and Au has been shown to reduce the WF and increase QE while still retaining the robustness of the metal; nonetheless, the crystalline quality of the substrate is sacrificed [12]. Since low intrinsic emittance beams play an increasingly important role for some photoinjector driven applications, maintaining a high quality surface can be advantageous in avoiding emittance degradation [13].

A theoretical model by Nemeth et al. [2] describes the density functional theory (DFT) simulation of a multilayered structure MgO/Ag(0 0 1)/MgO in the configuration of 4 ML of Ag(0 0 1) flanked by n ML of MgO. This model indicates that 2 or 3 ML of MgO can reduce the emittance of an electron beam, because the surface band structure exhibits a narrowing of the range of surface-parallel momenta near the Γ-point neighboring the Fermi level. Since the surface-parallel momenta of the electrons are conserved in prompt emission, the angular spread of emitted electrons (and so the intrinsic beam emittance) is reduced. In addition, this and another similar model [5] predict a WF drop of 1–1.5 eV from that of 4 ML of Ag(0 0 1). Measurements using atomic and Kelvin probe force microscopies (AFM and KPFM) show that even a one layer deposition of MgO on a Ag(0 0 1) single crystal substrate produces a WF drop of 1.1 eV (1 ML) and 1.4 eV (2 ML) from that of the bare substrate [3,4]. Recently, an effort to quantify the effect on emittance of a single 4 ML coating of MgO on Ag(0 0 1) was carried out using Angle Resolved Photoelectron Spectroscopy (ARPES) by Droubay

* Corresponding author. Tel.: +1 6302529708; fax: +1 6302520358.
E-mail address: terryj@iit.edu (J. Terry).

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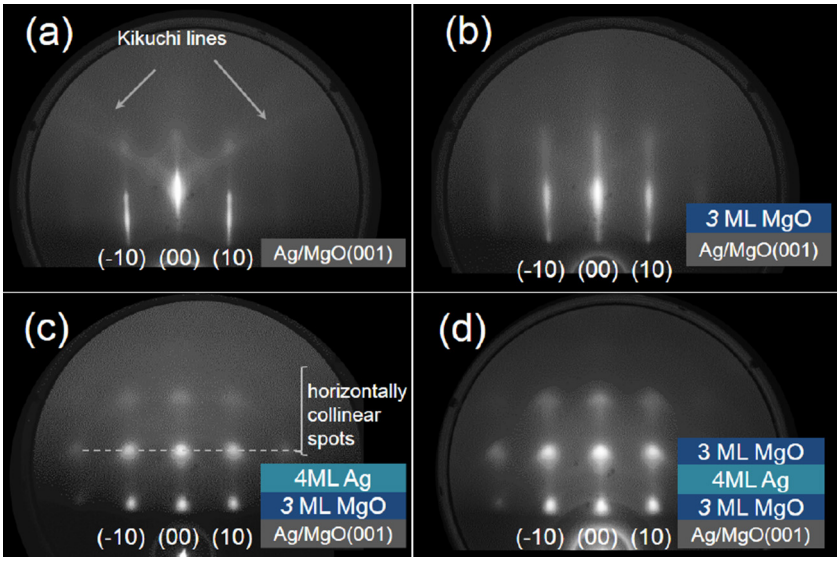

Fig. 1. RHEED patterns during deposition of multilayer $n = 3$ ML: (a) Ag/MgO(0 0 1) support, (b)–(d) progression of multilayer growth.

et al. [14]. The photon energy was not optimized, so the emittance of the coated sample was larger than from the substrate alone. However, an enhancement of the photoemission intensity at the Γ-point near the Fermi level was observed for the coated versus the uncoated Ag(0 0 1) substrate.

Here the dependence of emission behavior on the thickness of the MgO layers of multilayered MgO/Ag/MgO structures was examined. The high quality multilayer films were fabricated using pulsed laser deposition. Reflection High-Energy Electron Diffraction (RHEED) [15–17] and Photoelectron Spectroscopy (PES) [18,19] were used to monitor the formation of the highly oriented, crystalline, multilayer films, and verify the chemical structure of each. The Kelvin Probe technique was used to determine any changes in the work function as a function of the thickness of the MgO layers. QE was measured in vacuum by grounding the samples, illuminating them with UV light and collecting the photocurrent using an anode at 450 V. ARPES was used to measure the angular emission profile of the multilayer $n = 3$.

2. Materials and methods

Pulsed Laser Deposition (PLD), a state-of-the-art ultra-high vacuum film synthesis technique [20–23], was used for growth of all films described here. The laser fluence for all instances was 5–6 J/cm^2. Substrate heating was performed using a continuous wave CO_2 laser (10.6 μm) that impinges on the back of the substrate through a zinc-selenide window. Film growth monitoring was carried out using in-situ RHEED. The deposition rate was calibrated before growth of each film using a quartz crystal microbalance (QCM) with an uncertainty of less than 10% of the film thickness. Thickness measurements were independently verified using ellipsometry showing a small average discrepancy of ±0.03 Å. We observed that the roughness of the deposited surfaces was reproducibly dependent on the substrate temperature during deposition. The surfaces of the films for different layer thicknesses were consistent in quality from sample to sample.

3. Results and discussion

3.1. Synthesis and structural characterization

The multilayer MgO/Ag/MgO structures, in the (0 0 1) orientation, were fabricated on smooth epitaxial 40 nm Ag buffer films deposited on MgO(0 0 1) in order to substitute more expensive commercially available single crystal metallic substrates. Multilayers consisted of a 4 ML Ag interlayer flanked by $n = 2$, 3, 4 MgO monolayers, where the nominal monolayer thicknesses (i.e. half of the lattice parameter) are: 2.106 Å and 2.045 Å for (0 0 1) MgO and Ag, respectively [2]. The deposition temperature was 130 °C and the chamber base pressure was ~5×10^{-9} Torr for all films.

After growth, the films were initially characterized in order to study their structural properties. During growth the films were monitored in situ using RHEED in order to examine their crystalline structure. An example of multilayer evolution (for $n = 3$ ML) during growth, examined by RHEED, is shown in Fig. 1. Fig. 1(a) shows the pattern of the Ag/MgO(0 0 1) growth platform. Thin rods and Kikuchi lines as well as the absence of transmission-like features indicate that the film has become a terraced, 2-dimensional surface [6,15]. The RHEED system was calibrated using the pattern from MgO(0 0 1) (not shown). The calibration showed that the lattice constant for the Ag/MgO(0 0 1) pattern was 4.01 ± 0.03 Å, ~2% smaller than the nominal value of 4.09 Å for bulk Ag(0 0 1) [2]. Fig. 1(b)–(d) shows RHEED patterns of the sequential deposition of MgO (3 ML), Ag (4 ML), MgO (3 ML) on Ag/MgO(0 0 1). Epitaxial growth is manifested by the absence of polycrystalline features (rings and/or curved rods) [15–17]. The appearance of distinctive horizontally collinear features is indicative of transmission diffraction from 3D surface features [17], thus it is clear that the films grew in the form of highly ordered epitaxial islands. All patterns were taken along the [1 0 0] azimuth of MgO(0 0 1).

The multilayer surfaces were then imaged by STM in order to confirm the results from RHEED. Fig. 2 depicts the STM images corresponding to the multilayer $n = 3$ ML described in Fig. 1. Fig. 2(a) confirms the highly ordered, terraced surface finish of the Ag/MgO(0 0 1) growth platform, and Fig. 2(b)–(d) shows the formation of the multilayer as epitaxial nano-islands. The average and rms surface roughness for the Ag/MgO(0 0 1) support were measured to be 0.42 ± 0.03 nm and 0.52 ± 0.04 nm, respectively. The average and rms surface roughness for the multilayer surface $n = 3$ ML were measured to be 0.56 ± 0.03 nm and 0.71 ± 0.03 nm, respectively. Roughness was sampled over 100×100 nm^2 areas.

In addition to the characterization of the physical structure by RHEED and STM, ex-situ PES was used to track the chemical structure of the multilayered films at different stages of deposition. Fig. 3 shows Al K_α PES spectra of the evolution of deposition for the