



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



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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(001)/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.

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## 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(001)/MgO in the configuration of 4 ML of Ag(001) 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  $\Gamma$ -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(001). Measurements using atomic and Kelvin probe force microscopies (AFM and KPFM) show that even a one layer deposition of MgO on a Ag(001) 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(001) was carried out using Angle Resolved Photoelectron Spectroscopy (ARPES) by Droubay

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