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Editorial

Special Issue "Pulsed Laser Deposition of Thin Films: Recent Advances and Challenge"

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Since its introduction several decades ago, Pulsed Laser Deposition (PLD), has proved to be a powerful technique for synthesizing a broad spectrum of functional thin films. Indeed, with respect to other conventional physical vapor deposition techniques, PLD has several advantages, including but not limited to control of stoichiometry (especially for materials with complex composition), adherent coatings, easy-to-obtain multi-layered thin films and combinatorial maps, good versatility of experimental design, and morphology and crystallinity control. Moreover, it is possible to tailor the composition–structure–properties relationship of the coatings by adjusting the deposition conditions.

Matrix-assisted pulsed laser evaporation (MAPLE) is a complementary deposition technique derived from PLD, extensively explored for the deposition of thin organic nanoparticle and composite coatings. High experimental versatility allows for the synthesis of delicate compounds, such as proteins or polymers, without impeding the stability of their functional characteristics. Although initially designed for polymers, MAPLE has evolved to be used for various applications in biomimetic coatings, energy, sensing, wearable electronics, and photonic devices.

This Special Issue presents a topical collection devoted to recent advances and challenges in the field of pulsed laser deposition of thin films. Both PLD and MAPLE techniques are explored for specific applications in photovoltaic cells [1–3], optics and microelectronics [4,5], quantum materials [6], and wearable pressure sensors [7]. The Langmuir probe technique, as an in situ and real-time plasma characterization during the synthesis of the coatings, was reviewed in [8]. Additionally, laser coating via state-of-the-art additive manufacturing was reported in a comprehensive review [9]. This Special Issue contains three reviews and six research papers authored by leading scientists in the field.

In the context of the global energy crisis, the enormous potential of organic photovoltaic cells (OPC) is generally accepted in terms of producing cheaper and cleaner energy. However, the improvement of OPC performance is still a challenge that strongly depends on the materials and processing techniques used. Binary and ternary organic bulk heterojunctions based on zinc phthalocyanine were fabricated by MAPLE [1]. An increase in the open-circuit voltage is reported for the ternary blend in comparison with the binary blend, revealing that MAPLE coatings containing organic bulk heterojunctions can find applications in the field of photovoltaic device manufacturing.

An overview of the organic thin films deposited by MAPLE for photovoltaic cell applications is introduced in a comprehensive review paper [2]. The authors provided evidence for the recent advances obtained in the deposition of organic layers consisting of small molecule compounds, oligomers, and polymers using MAPLE-based deposition techniques (MAPLE, RIR-MAPLE and emulsion-based RIR-MAPLE). Moreover, the influence of laser processing parameters on the final properties of the layers is highlighted in order to discover new strategies for enhancing the OPC performances.

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Pulsed Laser Deposition was employed in [3] to synthesize active semiconductor TiO_2 bilayers, as photoanode structures in the fabrication of dye-sensitized solar cells. The layers consist of a thin, compact buffer and a thick, nanostructured layer deposited in different experimental conditions with a view to finding the optimum deposition regime. Thus, PLD thin film synthesis was conducted in He, N_2, O_2, and Ar atmospheres, followed by several physical–chemical investigations. The best electro-optical performance was obtained for photovoltaic structures based on TiO_2 coatings grown in He, and related to the greater roughness of the coatings and thus to a more efficient and deeper penetration of the dye inside the nanostructured layers.

Complex materials such as hafnium and aluminum silicate thin films, which have controlled stoichiometry and thickness as well as smooth surface features, are of great interest in optics and electronics. Pulsed Laser Deposition in optimized processing conditions, followed by a broad spectrum of physical–chemical analyses, has revealed properties suitable to use such coatings for laser beam delivery/handling systems [4].

During the last few decades, miniaturization has become a leading trend in microelectronics, while the development of buried oxide layer structures is one of the main improvements for low power electronics. The surface topography of Si/TiO_2 stacked layers on silicon substrates deposited by PLD as a function of target-to-substrate distance was investigated in [5]. Atomic force microscopy data analysis based on power spectral density (PSD) and the roughness-length scale (RLS) functions were used to quantify the results. It is shown that the relative RLS based on PSD function, used for AFM data analysis, quantitatively reveals the evolution of the surface profile on the top layer of the $Si/TiO_2/(100)$ Si-stacked films after their consecutive depositions, in relation to both the substrate and the target-to-substrate distance [5].

Today, the synthesis and investigation of ultrathin coatings is a challenge for all PVD technologies. In the case of quantum materials, one needs in situ control of the growth phenomena and real-time surface quality control of the films in order to assess the final intrinsic properties. Epitaxial FeSe thin films were grown by PLD on CaF_2 (001) substrates [6]. The thin films synthesis protocols were fine-tuned by optimizing target-to-substrate distance and ablation frequency. It is concluded that in situ analyses enable broad perspectives for the PLD deposition of various heterostructures in the global field of material sciences.

Piezoelectric hybrid heterostructures of $Ba(Ti_{0.8}Zr_{0.2})O_3$-$x(Ba_{0.7}Ca_{0.3})TiO_3$ (BCTZ) and polyvinylidene difluoride (PVDF) were obtained by laser techniques (PLD and MAPLE) on solid substrates as viable tools for the fabrication of wearable pressure sensors [7]. High values of the piezoelectric coefficient d33 and capacitance, as well as low dielectric losses, were achieved in this study. The possibility of integrating both ceramic and polymer piezoelectric materials into a single structure is demonstrated. As a clear advantage, it is stated that various multilayer heterostructures can be obtain without influencing the functional properties of the previous layer, as in the case of sol–gel or other chemical synthesis approaches.

Researchers generally accept the need for real-time in situ diagnostics and feedback loops for the systematic, semi-industrial implementation of PLD and other PVD techniques [8]. Indeed, in the case of PLD, transient plasma characteristics make quantitative plume analyses difficult, due to fast time- and space-changing parameters, rapid evolution, and complex compositional chemistry. The Langmuir probe technique proved to be a valuable tool for plasma diagnostics in the case of laser ablation and PLD. The review paper in [8] provides a critical insight on the limits and understanding of the technique as a foundation for attaining its full potential and implementation in industrial applications.

In material sciences, ceramics and ceramic-reinforced metal matrix composites have shown excellent properties such as high wear resistance, excellent chemical inertness, and exceptional properties at elevated temperatures [9]. Such characteristics make them suitable for several applications in biomedical fields, aerospace, electronics, and other high-tech applications. However, due to high fabrication limitations using conventional manufacturing methods (high costs and energy consumption), laser additive manufacturing (LAM) techniques, with high-power laser beams, have been developed and extensively employed for processing ceramics and ceramic-reinforced CMMC-based coatings. In respect to other LAM processes, laser melting deposition (LMD) excels in several aspects, such as high coating efficiency and lower labor costs. Nevertheless, difficulties such as poor bonding between coating and substrate, cracking, and reduced toughness are still relevant in some LMD coatings. In this review article [9], the authors overview the recent developments in the LMD of ceramics and CMMC-based coatings. Distinct facts and possible solutions, along with developmental challenges, are introduced, discussed and summarized in view of implementing this technology for current industrial use.

Thin and ultra-thin films have already proven to be at the forefront of various technological advances and will continue to impact our future by the discovery of new composite materials or innovative properties. A broad field of materials with complex stoichiometry can be deposited by PLD and MAPLE, as demonstrated in this Special Issue. We consider that novel, multifunctional coatings will broaden the spectrum of applications towards emerging cutting-edge technologies.

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