Molybdenum trioxide thin films for infrared Photonics

R. Macaluso[1], F. V. Lupo[1], M. Mercurio[2], D. Ceneda[2], B. Megna[1], D. Persano Adorno[3], M. Mosca[1], I. Crupi[1], M. Centini[2] and M. C. Larciprete[2]

[1] Department of Engineering, University of Palermo, Palermo, 90128, Italy
[2] Dipartimento di Scienze di Base ed Applicate per l'Ingegneria, Sapienza Università di Roma, Roma, 00161, Italy
[3] Department of Physics and Chemistry "E. Segré", University of Palermo, Palermo, 90128, Italy

roberto.macaluso@unipa.it

Abstract. Alpha-phase molybdenum trioxide (α-MoO_3) has attracted increasing attention in recent years due to its exciting physical properties which offer new opportunities for realizing Photonic devices with enhanced or novel functionalities. In fact, besides its strong anisotropy related to optical phonons, which allows polarization control and rotation, MoO_3 supports strong field localization by the excitation of surface waves called surface phonon polaritons (SPhPs), achieved through the coupling of the electromagnetic field with lattice vibrations. In this work we report on fabrication and structural, morphological, and optical characterization of polycrystalline α-MoO_3 thin films synthesized by pulsed laser deposition at 400 °C. Raman spectroscopy measurements revealed the polycrystalline single α-phase nature of the deposited MoO_3 films, while scanning electron microscope analysis showed a random grain distribution, which prevents optical anisotropy at normal incidence. Moreover, optical measurements in the mid-infrared showed an enhanced polarization-tuneable reflection peak at 1005 cm^{-1} both in amplitude and frequency, and a polarization-independent perfect absorption behaviour at 979 cm^{-1}, when 15° angular incidence is used. These interesting spectral properties, not least having preserved the typical strong dispersion related to the phononic response of α-MoO_3 flakes, indicate that pulsed laser deposition is a valuable technique to obtain large area and good quality α-MoO_3 thin films for infrared Photonic applications, including mid-infrared metamaterial devices for sensing applications.

Keywords: Molybdenum trioxide, polar materials, pulsed laser deposition, Mid-IR Photonics, phonon resonance.

1 Introduction

Alpha-phase molybdenum trioxide (α-MoO_3), is a polar dielectric material, characterized by surface phonon polariton (SPhP) resonances across three distinct wavelength bands (10 – 20 μm) for the three orthogonal crystallographic directions [1]. Because of these properties, novel nanophotonic devices based on α-MoO_3 have been proposed, including broadband absorbers, high quality factor polariton resonators, and polarization-sensitive optical devices. A metamaterial approach, based on the random micro structuration of α-MoO_3 with subwavelength dielectric elements has also been proposed. The latter could allow a large design versatility, tunability and hybridization of polariton modes, essential for sensing applications in the mid infrared. Despite the huge potential of this promising material, the development of a novel, highly versatile and compact α-MoO_3-based infrared (IR) Photonics platform is hampered by the lack of availability of high-quality films that can easily be integrated into a mature fabrication process. α-MoO_3 for IR Photonics has been so far mostly employed in the form of mechanically exfoliated flakes. Although flakes allow exciting results in terms of hyperbolic phonon polariton excitation along *x* and *y* directions, there are several practical limitations to the wide adaption of flakes geometries, not least their micrometer-scale dimensions and their irregular shape which can prevent a good propagation of SPhPs.
Pulsed laser deposition (PLD), due to its large versatility and low-cost, has proven to be a very effective technique to grow metal oxide thin films, including α-MoO_3 [1]. Compared with the exfoliation technique, it allows depositing large area MoO_3 films, which can be much more easily handled and integrated into a multilayer structure. In this work, we report on fabrication, structural, morphological and IR optical characterization of α-MoO_3 thin films synthesized by PLD.

2 Experimental

The employed PLD system uses a Q-switched Nd:YAG laser (λ = 355 nm, pulse width 6 ns) [3]. MoO_3 thin films were deposited onto fused silica substrates at 400 °C and 10^{-1} mbar O_2 pressure. Structural characterization of the films was assessed by Raman spectroscopy, while morphological characterization was carried out by scanning electron microscopy (SEM). Optical characterization was performed by using an FT-IR interferometer in the 7000 - 400 cm^{-1} frequency range.

3 Results and discussion

Fig. 1(left) reveals that the deposited MoO_3 film exhibits Raman peaks ascribable to the α-phase of MoO_3 [2]. In Fig. 1(right), the SEM top view of the film is reported. It is evident that the material is composed of randomly oriented grains with a layered structure, suggesting that the measured optical properties will not show strong anisotropic effects. Polarization-dependent FT-IR reflection measurements carried out at

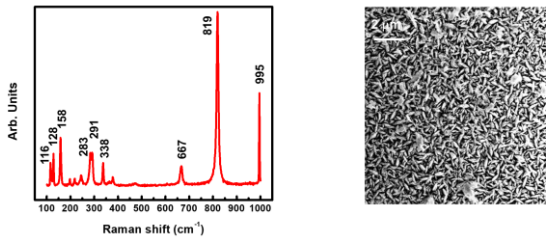

Fig. 1. Raman spectrum of a α-MoO_3 thin film deposited onto fused silica at 400 °C and 10^{-1} mbar oxygen pressure (left) and representative SEM top view (right). The scale bar is 2 μm.

15° and 45° incidence angles (Fig. 2) showed an enhanced tunability *vs* light polarization angle at around 1005 cm^{-1}, much more evident at 45° incidence angle. The latter case highlights also a slight frequency shift from 1002 cm^{-1} to 1005 cm^{-1} with different polarization angle. On the other hand, a polarization-independent perfect absorption behavior can be observed at 979 cm^{-1} for 15° incidence angle.

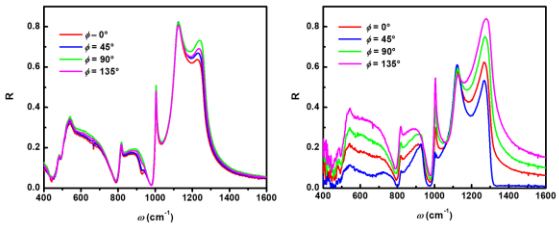

Fig. 2. Polarization-dependent reflection FT-IR spectra measured at 15° (left) and 45° (right) incidence angle. ϕ stands for polarization angle.

In conclusion, polycrystalline α-MoO_3 films with enhanced phonon polaritons resonances were deposited at 400 °C by PLD. The high reflectance dynamic range and the sharp resonance found may have great potential for the realization of mid-IR metamaterial devices for sensing applications, as well as for narrow-band detection.

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