NUCLEAAR

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I. SUMMARY

NUCLEAAR is a project funded by the INFN Vth Committee, which addresses scientific and technological challenges aimed to advances research frontiers on accelerators. The RD was oriented to technological advancements useful for future colliders and next generation of accelerators after LHC, a relevant issue of the European Particle Physics Strategy Form. In this context the aim of the NUCLEAAR project is the investigation of novel coatings to improve the performance of copper cavities. Actually, to design more powerful and compact accelerators, it is necessary to enhance the electromagnetic fields at which accelerating devices operate. The present Cu-based technology is limited by breakdown phenomena that unavoidably damage the surface of RF cavities operating for long time at electrical gradients $> 100 - 200\,MV/m$. Additional information are available at the URL: http://w3.lnf.infn.it/ricerca/ricerca-tecnologica/nucleaar/.

II. ACTIVITY

In spite the pandemic that seriously affected all experimental activities in the last couple of years, we confirm that the milestones of the experiments have been substantially reached. Indeed, for what concerns the two final milestones:

- Preparation of conductive films of TM oxides on curved copper surfaces
- Realization of a small prototype of a radiofrequency cavity with TM oxide coating

The first has been reached at 100%, while for the second we consider a percentage of $80 - 90\%$. In fact, in 2021 we brazed a small prototype with the first evaporations on curved surfaces and we completed the evaporation of the surfaces of the second, about 10 cm long. Unfortunately, at the end of 2021 it was not possible to complete the order to assemble the second prototype, which will only be available in the first months of 2022. The results obtained allowed us to prepare an internal note in which the entire process is described: from the preparation of the substrates up to the assembly of a cylindrical copper cavity 10 cm long with the internal surface coated by molybdenum oxide, TIG welded. The text describes the critical issues and the technological advantages of the process we have imagined and implemented in the framework of the NUCLEAAR project. To achieve all objectives described above, we completed the installation of a new heater in the existing evaporation chamber[1, 2] (see previous activity reports) and a translator with a sideshift stroke of $10\,cm$. Starting from January 2021 the chamber was reassembled and the sample heating setup was installed in the upper section. The chamber reached a limit vacuum of $8 \cdot 10^6\,mbar$. Tests carried out on silicon substrates were calibrated for the thickness using the AFM technique (Camerino University). Copper substrates with a roughness of 25 nm were available at the end of May 2021 and evaporation on Cu samples started. At the end of June the heating setup was installed and the ultimate measured vacuum of this setup is $1 \cdot 10^5\,mbar$ at a maximum temperature of $600°C$. While the heating element is capable to reach higher temperatures, tests were limited to $600°C$ without cooling the copper feedthroughs. Finally, a dedicated sample holder with four stations was prepared. The reflectivity measurements of the MoO_3 coated on copper have provided very interesting results which will be the subject of a publication, which is in progress. With the assembled system we are trying to successfully evaporate other oxides as well and to avoid contamination different crucibles were used. In 2021, the collaboration with English groups (Cambridge university, Cranfield university and Nottingham university) was successfully established. On the first thin films ($< 100nm$) obtained with a laser technique (PLD) [3] we have already carried out optical and Raman measurements and electrical characterizations. In the second half of 2021, other samples were made available and we characterized them as the previous ones. A joint publication on these PLD film is also under preparation. Always in 2021 low temperature transport measurements with the collaboration of the INFN unit of Naples, and damage tests at high electric fields [$\sim GV/m$] were carried out on both the films grown in Frascati and those produced by the English teams using the THz radiation of the FEL THz ISIR at OSAKA [4, 5]. The analysis is in progress on both datasets and we plan to publish the results in the near future. The collaboration with Seoul National University for the growth of MoO_3 crystals has been also very successful. The availability of single crystals has allowed to have MoO_3 "flakes" for unique studies of metal/MoO_3 interfaces that have been the subject of a thesis by a student of the University of Camerino[6]. Finally, in addition to the damage tests at high electric fields [$\sim GV/m$] using the THz radiation of the

FEL at Osaka, at the end of 2021 also the TERA source in Rome was available and the first tests have been carried out. The next experiments of damage will be carried out with the TERA source and we expect to reproduce the effects previously observed at Osaka on copper coated with MoO_3.

A. THz, IR and UV spectroscopy

Several experiments of THz spectroscopy werr performed using the THz-Time Domain Spectroscopy system for linear measurements. The electro-optical generation of the THz signal is achieved by the excitation of carriers in a GaAs layer inside a photoconductive antenna. The laser optical pulse of 780 nm is generated by a Toptica FemtoFiber Pro, with an 80 MHz repetition rate and pulse width $< 100 fs$. The Hamamatsu photoconductive antennas are used both for the THz generation and detection. The sample and THz setup are inside a closed chamber filled with nitrogen in order to eliminate the water vapors that affect the THz spectra. Measurements were performed with a reflectance setup composed by two gold mirrors that focus the signal on the sample and collect the reflected component to the detector. Specular reflectance IR spectroscopy were also performed using a Hyperion 3000 Bruker FT-IR Microscope paired to a Vertex 70v Bruker FT-IR interferometer. The microscope setup allows to measure from 400 to 8000 cm^{-1} collecting the MIR part of the spectra. To measure the FIR region down to 30 cm^{-1} a reflectance setup was used in a vacuum environment inside the sample compartment of the Vertex interferometer. Reflectance spectra show a good THz reflectance of all measured samples. A plasma edge appears at the 200 nm thick film, which can be related to a decrease in conductivity of the film. As in the THz characterizations, these experiments show the good reflectivity of the layers and a good metallic response of the Cu/MoO_3 system in the IR region. To extend the transmittance measurements in the visible and in the ultraviolet region, we used the spectrophotometer Jasco V-770. It incorporates two gratings and two detectors to fully cover the wavelength range from 190 to 3200 nm (UV-VIS-NIR). It features a single monochromator Czerny-Turner with two light sources: a Halogen lamp, which works from 2500 nm until the exchange wavelength, and a Deuterium lamp, which works from the latter wavelength until 190 nm. The Reflectance of the samples in the NIR-UV range exhibits a band gap lower (1.8 eV) for the 200 nm film respect to the 100 nm film (3 eV). This behavior could be related to a more disordered state in the thicker film[7].

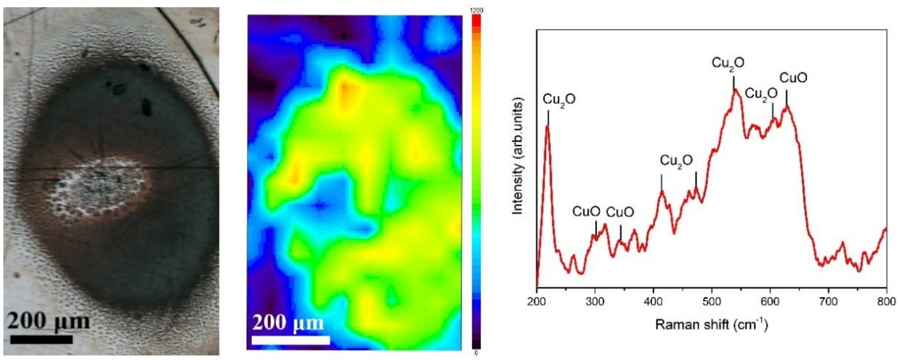

FIG. 1 : Optical image (left) of the copper region irradiated by 5000 THz shots at the incidence angle of $40°$. Raman spectra (right) and map (center) of the Cu_2O in the damaged area.

In order to test the resistance of the MoO_3 coating at high intensity Electric Fields we performed damage tests at the ISIR Free Electron Laser of the Osaka University. This FEL generates a coherent EM radiation at 3 THz (100 m) [4, 5]. In this breakdown experiments, the beam was focused on the surface to generate in a controlled and repetitive way irradiations with electric fields up to 4 GV/m. Several samples were characterized as a function of the Intensity, controlling the number of pulses, from 100 to 1000 pulses at the incidence angle of 40° and controlling also the beam energy with attenuators from 10% to 100% of the macro-pulse energy. We tested also on some samples the s polarizations: at $0°$ and $40°$ and the damage vs. the angle (p-polarization) at $0°$, $20°$, $40°$ and $60°$. The analysis of the damaged area and of the coating resistance has been performed using IR and Raman spectroscopy [Fig. 1].

B. MoO_x on copper

Various MoO_x samples deposited on copper have been investigated using Raman spectroscopy. In Figure 2 we show the optical image of the sample R26 deposited using the PLD with a nominal thickness of 500 nm. The presence

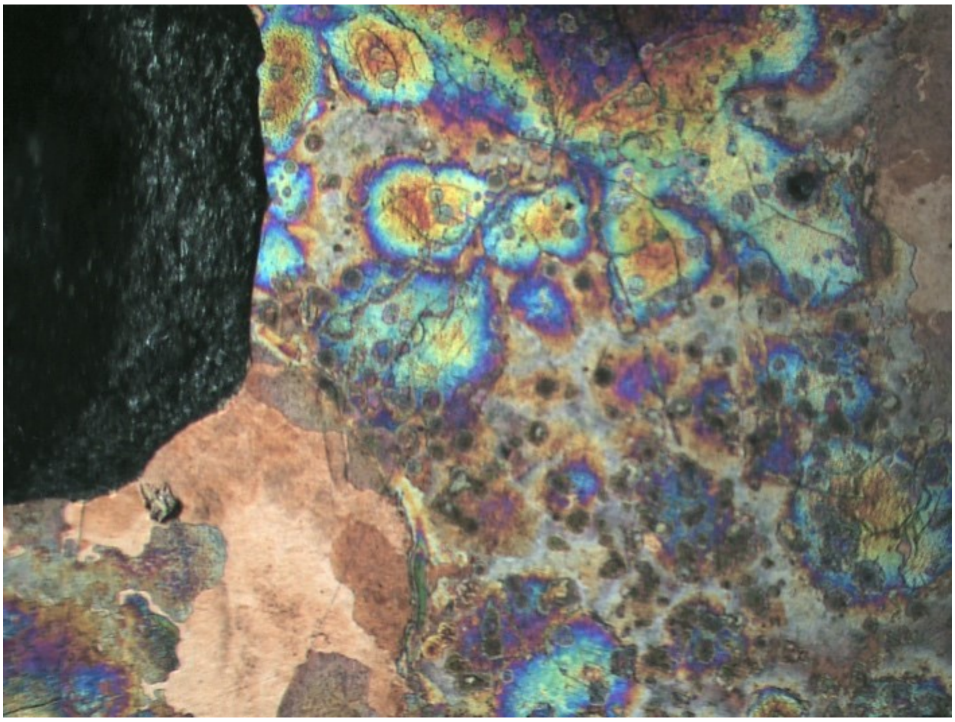

FIG. 2 : Optical image ($500x300\mu m$) of the sample R26 acquired with a x5 objective

of iridescent puddles suggests that this sample is not crystalline MoO_3 and the presence of regions with different stoichiometry or at least different amounts of defects that cause the different colours. The Raman spectrum confirms this hypothesis because no features belonging to MoO_3 can be identified while the presence of the broad peak suggests that the material is amorphous, with the coexistence of multiple phases randomly distributed. A comparison with the Raman spectrum of an amorphous MoO_3 film (Figure 3) deposited using the PVD method shows a shift of about 15 cm-1 in the peak position. This shift confirms the coexistence of multiple phases in the samples deposited with PLD. In the right panel in Figure 3 the most intense peak around 920-950 cm-1 does not match the pattern of the MoO_3 peaks, nor those of other Mo oxides known[8]. The analysis of these samples confirms the coexistence of multiple

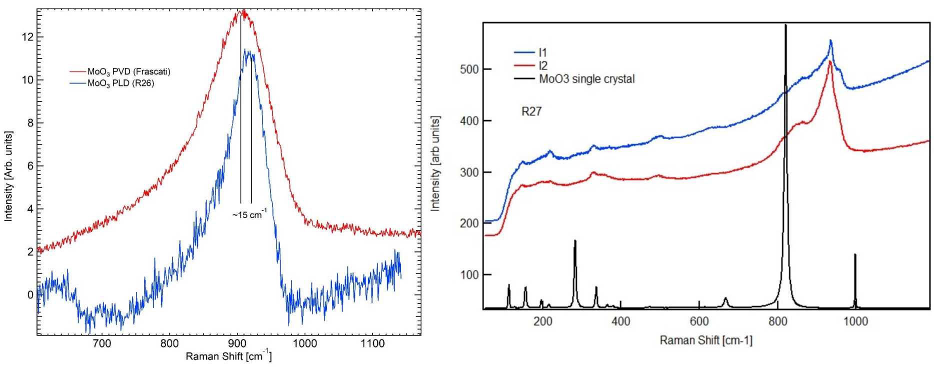

FIG. 3 : (Left) Comparison between the Raman spectrum of a MoO_3 sample deposited using the physical vapour deposition and the sample R26 with PLD. (Right) Raman spectra of the sample R27 in two different locations and of the MoO_3 single crystal.

phases of molybdenum hydroxide and other organo-metal oxides of molybdenum. As seen above, single crystalline samples growth at the Seoul university, have been investigated using Raman spectroscopy. The preliminary results report an anisotropic behaviour of the Raman spectra. As reported by Wang and co-workers[9], the intensity of the Raman peak at 820 cm-1 reaches the maximum, when the polarization vector is parallel to the a lattice parameter.

C. Cylindrical RF cavity coated prototype

In order to test the coating performance on a real cavity we assembled a copper-coated RF cavity with a cylindrical shape. A copper cylinder 100 mm long with an internal diameter of 60 mm and 80 mm of external diameter, was properly sectioned. Each section was diamond milled to obtain a roughness less than 10 nm (Figure 4).

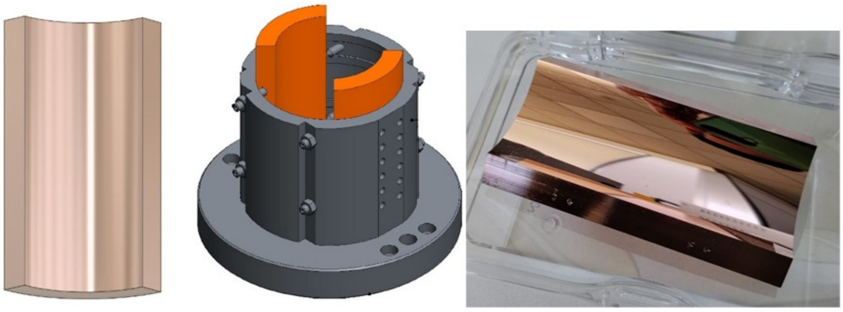

FIG. 4 : CAD scheme of the copper cylindrical sections and on the right the copper section before the evaporation.

III. ACKNOWLEDGEMENTS

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IV. PUBLICATIONS

- Francesco Paparoni Master Thesis, "Synthesis and Characterization of transition metal oxide films on metallic substrates" Camerino University 26 october 2021
- A. D'Elia, C. Grazioli, A. Cossaro, B. Li, C. Zou, J. Rezvani, N. Pinto, A. Marcelli, M. Coreno, Strain mediated Filling Control nature of the Metal-Insulator Transition of VO2 and electron correlation effects in Nanostructured films; Applied Surface Science, 540, 148341 (2021)
- A. D'Elia, S. J. Rezvani, N. Zema, F. Zuccaro, M. Fanetti, B. Belec, B. W. Li, C. W. Zou, C. Spezzani, M. Sacchi, A. Marcelli, M. Coreno; Stoichiometry and disorder influence over electronic structure in nanostructured VOx films; Journal of Nanoparticle Research, (2021).

V. ORAL CONTRIBUTIONS

- A.D'Elia, "Interplay of multiple degrees of freedom in strained VO2 films" 107°° Congresso della Società Italiana di Fisica (SIF), 13-17 September 2021.
- A.D'Elia, "Interplay of orbital hierarchy and metallicity in strained VO2 films" Bilateral Workshop on 3D Graphene and other 2D-3D Materials, Frascati, 25-26 November 2021
- S. Macis, "Thin conducting MoO_3 films on copper for technological applications: a new route for improved RF devices" Symposium of Quantum Materials for Quantum Technologies QMQT, Frascati 14 February 2022