Survey of metal oxides for coatings of ultra-stable optical cavities

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Abstract: A survey of the optical properties and mechanical loss of metal-oxide thin films is reported in a search for optimum materials for mirror coatings for gravitational wave detectors. Ta_2O_5 and GeO_2 doped with TiO_2 achieve the lowest absorption and mechanical loss with post-processing annealing.

1. Introduction

Metal-oxide multilayer dielectric coatings are engineered optical materials ubiquitous in optical systems. These structures consist of alternating layers of two materials of contrasting refractive index and a thickness that are selected to meet specific optical requirements. In the visible and near infrared the most widely used materials are Ta_2O_5 or HfO_2 (high index) and SiO_2 (low index). Multilayer stacks are grown by physical vapor deposition methods which have been optimized to realize thin films with superior optical quality. There are, however, applications such as the ultra-stable optical cavities of gravitational wave detectors, that place stringent demands on coating's optical performance and mechanical loss properties.

We report results of a survey of the optical properties and mechanical loss of doped and undoped metal oxide thin films, in a search for optimum materials for mirror coatings for gravitational wave detectors. Doping metal oxide alloys provide flexibility in the optimization of the optical response. It is shown that doping is also important to achieve an atomic configuration in the amorphous materials that is conducive to the lowest mechanical loss and hence reduced thermal noise in the coatings, which limits the sensitivity of gravitational wave detectors.

Coating thermal noise arises from thermally driven random fluctuations in the amorphous thin films that contribute to elastic loss. These losses can be quantified by measuring the mechanical loss angle ϕ of the materials. Mechanical energy dissipation is larger in the high refractive index layer compared to the low index layers which comprise the multilayer coating stack of the test masses of Advanced LIGO (aLIGO). Doping and post-deposition annealing have been shown to be most efficient in reducing the mechanical loss of the aLIGO coatings. Plans to further increase the sensitivity of gravitational wave detectors to probe further out into the universe make it necessary to identify metal oxides materials with reduced mechanical loss compared to the state of the art.

2. Results and Discussion

Metal oxide thin films were grown by ion beam sputtering and were post-deposition annealed in air at different temperatures until the onset of crystallization was observed. Their optical quality was assessed from ellipsometry and absorption loss measurements. The mechanical loss angle ϕ was determined from the analysis of the ring down of various resonant modes using a gentle nodal suspension (GeNS) system after each annealing step [1].

Figure 1a shows the coating loss angle at 1 kHz for TiO_2:Ta_2O_5 films with cation ratio of 0.27 and 0.53 as a function of annealing temperature. Doping Ta_2O_5 with TiO_2 reduces the loss angle, as has been previously reported [2]. Further reduction is obtained upon annealing. There is, however, a contrasting behavior at 600 °C, at which ϕ increases to 1.3×10^{-3} in the mixture with 0.53 Ti concentration, while it reaches its lowest value of $\phi = 0.28\times10^{-3}$ in the 0.27 Ti concentration sample. This is roughly a reduction of 40% in the loss angle compared to the annealed Ta_2O_5 film. At 600 °C the 0.27 Ti content mixture also achieves its lowest absorption loss at λ=1064 nm, 3.9 ppm for a film 200 nm thick [3]. The addition of 27% Ti causes significant morphological changes in the mixture.

Annealing promotes cation diffusion and the formation of a ternary compound, $TiTa_{18}O_{47}$[3]. In contrast, the 0.53 Ti content alloy phase separates, showing signs of crystallization of rutile TiO_2. Figure 1b summarizes results of loss angle for thin films of Ta_2O_5 doped with Nb, Zr, Hf, Al, Si, Zn, Sc and Y at a cation content of ~20% and at an annealing temperature 50 °C below crystallization. In all cases the addition of a dopant reduces the loss angle. The ZnO_2 doped Ta_2O_5 is the only mixture that achieves a value of the mechanical loss angle, $\phi=2.4\times10^{-4}$, lower than that of TiO_2 doped Ta_2O_5. The addition of Zn causes the materials to undergo structural modifications that lead to the formation of a ternary alloy upon annealing, as found in the case of TiO_2 doped Ta_2O_5. The optical properties are also modified with doping. The refractive index varies according to the law of mixtures. Doping Ta_2O_5 with Al, Si, and Zn reduces the absorption loss at λ=1064 nm from 2.5 ppm to about 1.3 ppm.

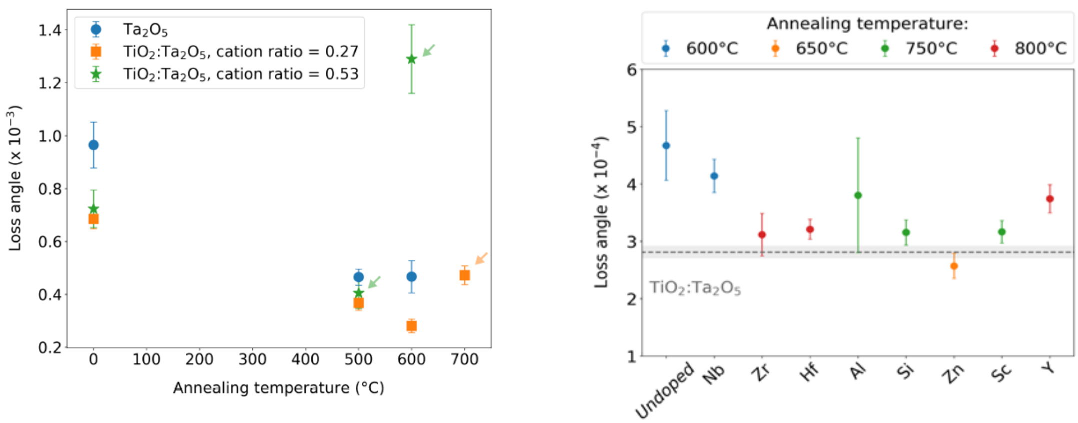

Fig. 1. (a) Coating loss angle at 1 kHz for TiO_2 doped Ta_2O_5 samples with Ti cation ratio of 0.27 and 0.53 as a function of annealing temperature. The arrows indicate crystallized films. (b) Coating loss angle for compounds of Ta_2O_5 mixed with other metals at a cation ratio of ~20% and after annealing to a temperature 50°C below crystallization.

We have also investigated doping of GeO_2 and SiO_2 with TiO_2. Both, SiO_2 and GeO_2 are glass forming materials which, in thin film form, have achieved the lowest mechanical loss among metal oxides, $\phi \sim 1\times10^{-4}$. However, their refractive index at λ=1064 nm is n=1.6 (GeO_2) and n=1.45 nm (SiO_2), which do not offer significant index contrast for the design of a GeO_2/SiO_2 multilayer. The refractive index can be tailored with the addition of Ti as shown in Fig. 2. With the addition of 44% content of Ti into GeO_2 the refractive index at λ=1064 nm increases to n=1.88. This coupled with an impressively low coating loss angle, $\phi=0.96\times10^{-4}$ and 3 ppm absorption loss at λ=1064 nm for a 216 nm thick film make TiO_2:GeO_2 well suited to replace TiO_2 doped Ta_2O_5 as the high index layer in interference coatings with reduced coating thermal noise for gravitational wave detectors.

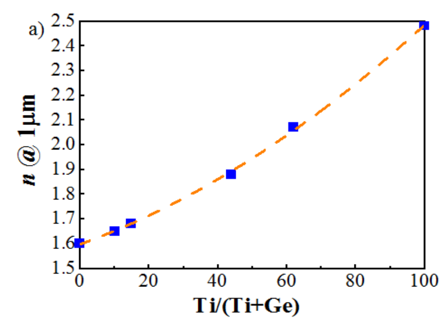

Figure 2. (a) Variation of the refractive index of TiO_2 doped GeO_2 at λ=1000 nm as a function of the Ti content.

3. References

[1] G. Vajente et al, A high throughput instrument to measure mechanical losses in thin film coatings, *Rev. Sci. Instruments* **88**, 073901 (2017).

[2] Gregory M Harry, et al. Titania-doped tantala/silica coatings for gravitational-wave detection. *Classical and Quantum Gravity*, 24(2):405, (2006).

[3] M. Fazio et al, Structure and morphology of low mechanical loss TiO_2-doped Ta_2O_5, *Opt. Mater. Express* **10**, 1687-1703 (2020).

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