The research on amorphous coatings for future GW detectors

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Abstract. The high mechanical losses of the multilayer reflecting coating deposited on the mirror surface account for the main contribution to the thermal noise, limiting the sensitivity in the mid-frequency region of the detection band of the future gravitational waves detectors. Several European laboratories of the Virgo Collaboration have joined their efforts to improve the coating mechanical performances. The research lines of this collaboration are all focused on amorphous coatings, which represent a viable solution for the future GW detector generations. The main target is to find a way to reduce by a factor three the mechanical losses of the coating for the next generation of room temperature operating detectors. Some activities are also meant to be relevant for cryogenic operations. The status of this collaborative work will be described as well as the latest results of the different research lines.

1. Introduction
The detection of gravitational wave (GW) signals by the two Advanced LIGO detectors [1] and by the Advanced Virgo detector [2], has been extremely important to test General Relativity theory in the strong field regime, but also to open a new window on the Universe with a completely new kind of astronomy. Improving the sensitivity of current detectors makes possible to increase the detection rate, to extract more information from each individual detected signal, and to improve the source localization capability of the detector network. The thermal noise of the main mirror surface coatings largely contributes to limit the design sensitivity of interferometric GW detectors, particularly in the middle part of their frequency band (30 Hz to 300 Hz). The mirror coating is made with a stack of alternate layers of low and high refractive index materials, forming a Bragg reflector. The coating layers are deposited through ion beam deposition technique on a silica substrate [3]. In current detectors, SiO$_2$ (silica) and TiO$_2$:Ta$_2$O$_5$ (titania doped tantala) are used as low index material and high index material, respectively. The final coating film presents a high reflective coefficient, and an extremely low optical absorption, below 1 ppm at 1064 nm [4].

Unfortunately from the mechanical point of view, TiO$_2$:Ta$_2$O$_5$ presents a much higher internal friction than SiO$_2$ [5]. As described by the fluctuation-dissipation theorem [6], friction is intrinsically related to thermal fluctuations, therefore high refractive index layers are responsible for most of the mirror total thermal noise. The aim of the Virgo Coating R&D (VCR&D) collaboration is to find new materials and new techniques to reduce the mirror thermal noise level in order to exploit the advancements developed in optical quantum noise.
2. Thermal noise in amorphous material and the VCR&D collaboration

In absence of a long range order in the atomic structure of amorphous solids, anelastic transitions of small group of atoms act as the main source of energy dissipation. These transitions can be modeled as the motion of a particle between the energy levels of a two level system (TLS) [7]. Energy dissipation occurs during the jump of the atoms from a potential well to the other. At low temperature the transitions are due to quantum tunneling while at higher temperature the process is thermally activated. Only transitions with a relaxation time of the same order of the period of the strain wave propagating in the material contribute to mechanical losses [8]. The barrier height of the TLS determines the characteristic relaxation time, thus only TLSs with a barrier height of 0.5 eV are relevant for the GW detectors frequency band at room temperature. Due to the lack of these 0.5 eV - TLSs, internal friction in SiO$_2$ is very low at room temperature, making SiO$_2$ the key material for current GW detectors [9]. On the other hand, lowering the temperature, as foreseen for future GW detectors, the number of relevant SiO$_2$ TLSs increases together with dissipation. The reduction of the total number of TLSs and the optimization of their distribution, are the guidelines of the VCR&D collaboration research. Eight Italian and two French laboratories are involved in VCR&D [10], all of these already members of the Virgo Collaboration. The activity of VCR&D includes coating synthesis, mechanical and optical characterizations, microscopical characterizations and the modeling of molecular structures. The main research lines cover the searching for new materials, the assessment of new post-deposition treatments, the understanding of the relationship between optical and mechanical losses and the possible improvement of the deposition techniques.

3. Highlights on research lines results

A number of interesting results are coming from the activities of the labs involved on the various research lines, some of which are briefly highlighted in the following. All the details can be found on referenced papers.

3.1. Updating of current coating mechanical characterization

SiO$_2$ is used as low refractive index material in all the current detectors. Ta$_2$O$_5$ is used in KAGRA as high index material, while TiO$_2$:Ta$_2$O$_5$ is used in Virgo and LIGO. New measurements of the mechanical properties of these materials were performed [11]. For each resonant mode of a coated SiO$_2$ thin disk, a measurement of the coating dilution factor (the fraction of energy stored inside the coating) was done by measuring the resonance frequency shift caused by the coating deposition. Mechanical properties as Young modulus, Poisson coefficient and density were extracted by matching the results of a finite-element analysis with the measured value of the dilution factors. Measured loss angles can be fitted by a frequency power law $\phi_{coat} = Af^B$. In the case of SiO$_2$, a term which quantifies spurious losses in the disk edge, is added to the power law, matching the mode branching [12].

3.2. Effect of the deposition parameters and the post-deposition annealing

Losses of the coating as deposited depend on deposition parameters, in particular on the deposition rate. Nevertheless for Ta$_2$O$_5$ a post deposition annealing (10h at 500 $^\circ$C) completely cancels any difference between depositions [13]. This sort of erasing effect is visible, even if less evident, in SiO$_2$ coating too. Post-deposition annealing is crucial in reducing coating losses. Raman spectroscopy revealed that the D$_2$ peak is reduced by the annealing process, in particular for SiO$_2$ this reduction appears to be large both for bulk and for film material. Since D$_2$ peak seems to be associated with the abundance of threefold rings of SiO$_2$ tetrahedral units [14], this result suggests that losses are tightly related to the population of this kind of microscopic molecular structures. For Ta$_2$O$_5$, the bigger reduction of losses and of the D$_2$ peak,
is reached after a 10h-500°C annealing, and no further reduction appears increasing time or rising temperature up to 600°C, when crystallization occurs.

3.3. Correlation between absorption and mechanical losses
The optical absorption coefficient curve of amorphous materials presents an exponential transition (Urbach tail) near the electronic band-gap. The broadening of this tail is related to the structural and thermal disorder of the material, and quantified by a parameter called Urbach energy. Recent research [15] showed a clear correlation between Urbach energy and mechanical losses. This correlation is visible for different kind of coatings (either pure Ta$_2$O$_5$ and Nb$_2$O$_5$ or mixed with Ta$_2$O$_5$) suggesting that this could be a general feature. This study also shows the effect of both annealing and doping, in decreasing Urbach energy, leading to more organized atomic structures, and reducing at the same time the mechanical losses. Urbach tails observation extends the spatial range of structural analysis in searching for correlations with mechanical losses, and can be used for a rapid estimation of the mechanical behavior of new materials.

3.4. Molecular Dynamics simulation
The Molecular Dynamics simulation of Dynamical Mechanical Spectroscopy (MD-DMS) has been developed to predict coating mechanical behavior[16], to improve the understanding of the dissipation mechanisms, and to rapidly evaluate new material performances. The MD-DMS approach is almost theory independent, where interatomic potential of the specific system is the only ingredient provided to the simulation. Mechanical losses are computed by the phase shift between an applied oscillating strain and the resulting stress. The simulation frequency range goes from GHz to THz, but an extrapolation in the acoustic band can be done by fitting with a frequency power law. Ta$_2$O$_5$ simulation gave promising outcomes and the extrapolated losses are in good agreement with the experimental measurements [17].

3.5. Non-oxide materials
In search for new low-losses materials, Si$_3$N$_4$ (silicon nitride) was shown to be a promising candidate for high refractive index layers [13]. Ion beam sputtered Si$_3$N$_4$ was annealed up to 900°C without evidence of crystallization, showing a loss angle about 3 times smaller than TiO$_2$:Ta$_2$O$_5$. Moreover an increase of the annealing temperature up to 900°C also decreases the dissipation of the SiO$_2$ layers, eventually improving the performance of the whole coating stack. The main open issue is that the extinction coefficient of Si$_3$N$_4$ does not yet meet the stringent absorption requirements for GW detectors.

Since fluorides are materials with the lowest index of refraction, studies are performed in particular on MgF$_2$ (magnesium fluoride) [18], for replacing SiO$_2$ in future cryogenic detectors. Currently room temperature measurements on the effects of annealing show a maximum reduction of dissipation with a 30 hours-400°C annealing, whereas a degradation appears at annealing temperatures higher than 500°C. Measurements at low temperature are foreseen in the next future.

3.6. Nano-layered coating deposition
Post-deposition annealing largely improves coating mechanical characteristics, but the maximum annealing temperature is limited by the beginning of the film crystallization. In nano-layered films the crystallization can be frustrated by the size of the layers, increasing the maximum allowed temperature [19]. For 10 - 100 nm scale TiO$_2$ layer, crystallization appears around 300°C, whereas going down with the layer thickness to nanometer scale, samples can be annealed up to 700°C with no evidence of crystallization [20] [21]. Within the VCR&D collaboration,
an ion assisted E-beam evaporation coater is devoted to nanolayered depositions to investigate the performances of different materials [22]. The first produced samples show good surface quality with a RMS of the surface roughness smaller than 1 nm. A possible optimization of the nanolayered stack is under investigation [23].

3.7. Metrology
Coating mechanical losses are computed by the difference between the value of the loss angle after and before the coating deposition. A very high level of accuracy is needed: the Gentle Nodal Suspension (GeNS) is the best loss angle measurement techniques available for this purposes [24]. In order to minimize spurious losses, a detailed measurement protocol must be established, and shared among all the laboratories. In many cases thin disk-shaped samples do not present a polished lateral surface. The effect of the resulting extra losses was effectively modeled [12], and the effect of a laser-polishing of the edges was assessed [25]. Since stability of substrate is mandatory, ageing effects are under investigation [25]. In case of thermo-elastic losses dominated substrate (silicon or sapphire thin disks), an experimental technique to estimate the thermo-elastic dissipation peak shift due to the coating deposition, is currently under study [26].

4. Conclusions
To improve the sensitivity of the future gravitational wave detectors, coating thermal noise must be reduced. Inside the VCR&D Collaboration several research lines are pursued in parallel by groups of laboratories, to maximize the likelihood of getting to a breakthrough. Research lines exploit both experimental characterizations and theoretical and computational modeling of the coating thermal noise sources. Many interesting results are coming out from the various research lines.

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