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Mirror Coatings for new generation Gravitational Wave Detectors(*)

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Summary. — Modern gravitational wave detectors are affected in sensitivity by thermal noise in the coating deposited on the mirror free masses of the interferometer. In the middle region of the frequency band (50-300 Hz), this constitutes the main limitation placed on the detector performance. The research activity on this front is focused on the exploration of new materials, deposition techniques, and post-deposition treatments, in order to minimise mechanical losses, optical absorption and scattering, for both components (high and low refractive index) of the multilayer stack constituting the Bragg reflector. Moreover, the possibility of employing cryogenic solutions for next-generation detectors, such as Einstein Telescope, introduces new requirements concerning the mechanical losses and the understanding of their underlying physical mechanisms, leading to the consideration of crystalline coatings alongside amorphous ones. We report here on the research activities within the Virgo Coating R&D Collaboration, with specific focus on the optimisation of mechanical and thermo-optical properties, and present an overview of the potential candidate materials being subject to investigation, including the specific technical and metrology challenges they raise.

Present-day gravitational wave detectors are constituted by a laser Michelson interferometer in which the light is reflected by free mirror masses [1-3]. In the middle region of the frequency band (50-300 Hz), the sensitivity is mainly limited by thermal noise of the coating deposited on the mirror surfaces. This carries important consequences on the physics to be observed, *e.g.*, the detection of coalescence phenomena of binary neutron stars and black holes is especially affected by noise within this frequency range. Therefore, a major goal to improve detector sensitivity lies in understanding and finding ways to reduce the coating thermal noise.

The mirror coatings consist of Bragg reflectors, namely, a multilayer stack of alternating low-refractive index and high-refractive index materials, deposited onto the mirror

(*) IFAE 2024 - "New Technologies" session substrate. With some simplifying assumptions, the power spectral density of this noise can be expressed as follows:

$$S(f) \propto \frac{k_B T}{f Y_{sub}} \frac{t_{coat}\phi_{coat}}{w^2} \tag{1}$$

where T is the temperature, f the frequency, Y_{sub} the substrate Young's modulus, w the laser spot size on the mirror, t_{coat} the total coating thickness. The material-dependent contribution is encapsulated by the loss angle ϕ_{coat}, which quantifies mechanical dissipation; indeed, from the fluctuation-dissipation theorem of condensed matter physics, it ensues that the thermal noise produced in a system is related to its capability for dissipating mechanical elastic energy.

Based on this, one can identify three main requirements that good coating materials must possess, in order to produce low thermal noise and also serve efficiently as a Bragg reflector:

1) low mechanical losses (ϕ_{coat}) for both materials

2) high contrast between refractive indexes of the two materials, which allows one to reduce the total thickness of the stack

3) low optical absorption and scattering: it is required that the total absorption in the stack be < 1 ppm. The band gap must be $>> 1.32$ eV to prevent transitions between valence and conduction bands at the laser wavelength of 1064 nm.

The state of the art in the detectors of current generation employs a stack of alternating amorphous silica (SiO_2) layers (as a low-index material) and titania-doped tantala (TiO_2:Ta_2O_5) layers (as a high-index material) [4]. With respect to requirement 1), the coating loss angle is in the order of 10^{-5} for silica, which is more than acceptable. For the high-index material, tantala (Ta_2O_5) would present a coating loss worse by one order of magnitude, in the range of 10^{-4}, a figure which is slightly ameliorated by the doping with titania (TiO_2). The optimal percentage for the doping (a 20%-80% mixture) has been determined by an empirical procedure of trial-and-error. Regarding the second requirement, on contrasting refractive indexes, for TiO_2:Ta_2O_5 the high-index value is $n_H = 2.09$, whereas for SiO_2 the low-index value is $n_L = 1.45$. Finally, as per the requirement on absorption, the extinction coefficient of each material in the stack is in the order of k $\approx 10^{-7}$, which is suitable for the detector.

Looking onwards from this starting point, one can define a list of objectives to be pursued for next generation coatings. In the first instance, the main goal is to reduce mechanical losses in the high-refractive index material; the conventional goal for ongoing research has been fixed to a reduction factor of at least 3. Concurrently, there has been a shift in the search for new materials by placing new emphasis on a deeper understanding of the underlying physical mechanisms driving the losses, thus moving away from a trial-and-error approach towards the identification a priori of potential candidate materials. This systematic approach is also expected to have beneficial repercussions for new generation detectors, such as Einstein Telescope, which will operate at cryogenic temperature in the low-frequency regime, and thus will necessitate a replacement of both coating materials, since fused silica dramatically increases its mechanical loss as the temperature is lowered. It is also important that this breakthrough on mechanical losses is attained without lowering performance on requirements 2) and 3) with respect to the current state-of-the-art.

To embark on this project, the Virgo Coating R&D Collaboration has been created, with the aim of meeting these targets for the future runs of O5 and post-O5 (Virgo_nEXT). The Collaboration is comprised of a network of universities and research centres located in Italy, France, Belgium, Netherlands and Japan. Among these, the Laboratoire for Matériaux Avancés (LMA) in Lyon possesses a facility for the deposition of coatings on substrates of large dimension, such as the Virgo mirrors.

The main techniques of experimental investigation available to the Collaboration can be broadly classified as follows:

- synthesis of new materials: study of deposition techniques, tuning of deposition parameters, characterisation of post-deposition treatments (thermal annealing)
- optical properties: measurement of optical absorption/extinction coefficient and refractive index (spectroscopic ellipsometry)
- mechanical dissipation properties: measurement of loss angle; measurements of density and elastic constants (Brillouin spectroscopy); numerical simulations (molecular dynamics simulations; Finite Element Analysis)
- microscopic structure: chemical composition and stoichiometry (XPS); incipient crystallisation (XRD; Raman spectroscopy); local molecular structures in amorphous materials (Raman spectroscopy); topology and surface composition (AFM; SEM)
- thermal and opto-thermical properties: optical path as a function of temperature (thermo-refractive measurements); measurement of the coefficient of linear thermal expansion (curvature measurement).

Taken together, these techniques offer a comprehensive picture of the relevant physics of a given material; they can also tie together in non-trivial ways, *e.g.*, with the discovery of how the optical properties, via the presence of Urbach tails, can offer a quick assessment of the expected mechanical behaviour [5].

1. – Amorphous Coatings

Turning now to the candidate materials under investigation, they can be divided into two large categories, amorphous and crystalline. Concerning amorphous materials [6], and analysing first the issue of mechanical dissipation, we can consider silica as an example to illustrate how the dissipation occurs. The silica building blocks are tetrahedra consisting of one silicon atom surrounded by four oxygens. Although the overall structure is disordered, the chains of tetrahedra connected at their vertexes can arrange themselves in locally ordered configurations; when two of these configurations are possible at a comparable energy level (Two-Level Systems, or TLS), the local ordering can then transform from one state to another by effect of thermal agitation. If we assume that the separation between meta-stable states is given by an activation energy barrier V, the characteristic time of these transitions is determined by the ratio between energy barrier and temperature. If this time coincides with the period of a propagating strain wave, then we have dissipation. This implies that the "critical" meta-stable states, which can give rise to mechanical dissipation, are those separated by a certain critical value of the energy barrier V, which is a function of temperature: it is situated at about 0.5 eV at room temperature, and its value decreases at cryogenic temperature. From this picture, it follows that, in order to minimise dissipation, there are chiefly two avenues available:

a) one can choose materials whose energy barrier is significantly lower than the critical value. These are “floppy” materials, which can easily transition locally from one meta-stable state to the other, such as silica (whose tetrahedra are only connected at their vertexes). This solution however is only viable at room temperature; at lower temperature, the critical value for V drops below 0.5 eV and therefore eventually dissipation is activated.

b) one can choose “stiff” materials. They can be materials for which V is larger than 0.5 eV, because the atoms possess a high coordination number with their neighbours, and therefore cannot easily alter their structure. Alternatively, their energy landscape can contain very few meta-stable states, so that transitions are intrinsically unlikely. Either way, stiff materials retain their low-dissipation properties also at low temperatures.

As for the specific materials under investigation, and the issues currently being addressed in their development, a brief overview can be given as follows. On the front of floppy materials, silica is already an established solution for the low-refractive index component of the stack; its mechanical dissipation as well as optical properties are well within the required specifications. A choice for the high-refractive index component is not equally obvious. Research has mostly been focused on oxides, since they are expected to guarantee a figure for optical absorption compatible with the requirements. The demands on mechanical dissipation, however, are not so easily met. Titania-doped germania (TiO_2:GeO_2) appears to be very promising as its mechanical dissipation should be an improvement on current values; it has been explored by the Center for Coatings Research within the LIGO Scientific Collaboration, and also developed at LMA. The main effort so far has been devoted to optimising the procedures for the deposition and the multilayer annealing.

On the front of stiff materials, amorphous silicon (a-Si) presents a low density of TLS states in its energy landscape. However, its energy bandgap is so low (≈ 1.12 eV) that optical absorption is dominant, at the customary laser wavelength of 1064 nm. Employing this material would therefore necessitate the increasing of the laser wavelength to 1550 nm. Another avenue being explored consists in nitrates and semiconductors whose atoms are arranged in covalent structures with high coordination number, and therefore are quite rigid with respect to transitions between meta-stable states (the energy barrier V is larger than 0.5 eV). In particular, Si_3N_4 has been tested for deposition at LMA since 2016; however, it is difficult to obtain stoichiometric coatings with no contamination, and as a result, at the present state the optical absorption is increased beyond expected values. As for amorphous semiconductors, the selection of candidate materials is being limited by their optical properties (the only materials with large bandgap are AlP, GaP, InP, and GaAs), as well as technical problems in deposition due to safety and toxicity concerns.

In conclusion, within the class of amorphous materials, the mechanisms underlying the physical dissipation are reasonably understood. From the technical side of things, the deposition of such coatings on a substrate of large dimensions is part of the current know-how and does not present particular challenges. The major limiting factors are instead to be found in optimising the stoichiometry and preventing contamination during the deposition, two issues that have yet to be addressed satisfactorily, and are likely the reason why amorphous materials have yet to be found to possess all of the desired properties they are supposed to, from a priori considerations.

2. – Crystalline Coatings

The second large category of materials is comprised of crystalline ones. In this respect, the situation is quite the opposite to the one mentioned above: it is not difficult to find materials that satisfy all of the required physical properties, but the difficulties arise in the technical aspects of large-scale deposition and substrate transfer (the stack must be removed from the crystalline substrate onto which it has been grown via molecular beam epitaxy, to be transferred onto the mirror). More specifically, regarding the mechanical dissipation, if the specimen is a good single crystal virtually deprived of defects (dislocations, grains, ...), we expect minimal intrinsic losses. In fact, in this case the contribution that becomes dominant for thermal noise is not brownian but rather thermo-optical, which is discussed in the following. Regarding the optical properties, the absence of disorder also plays a favourable role, by ensuring that conduction and valence electronic bands are well separated, with no intermediate levels within the gap that would facilitate absorption.

The candidate materials under consideration, at present, are GaAs/AlGaAs and crystalline oxides. AlGaAs has excellent mechanical properties (ϕ_{coat} in the order of 10^{-6}), so much so that it would allow a reduction of the laser beam spot, which is especially important given the technical challenges associated with large depositions for this category of materials. Research on it is being developed mostly within the LIGO Collaboration [7]. Crystalline oxides such as Cr_2O_3, Fe_2O_3, Ga_2O_3 [8] possess a corundum structure (space group 167), whose high level of symmetry may favour the growth of ordinate structures with no multiple crystallites and few defects. They also have the additional advantage of being grown on a sapphire substrate, since the lattice cell parameters between substrate and coating are well matched; this makes them a promising material also for Einstein Telescope, where sapphire is one of the candidate substrates being considered for the low-frequency, low-temperature branch of the interferometer. For all these materials, however, the state of the art is not yet at the level needed to ensure homogeneous deposition on a substrate on the diameter scale of the Virgo mirrors.

As anticipated, for crystalline materials the brownian thermal noise is significantly reduced, and consequently the thermo-optical thermal noise comes to the forefront. This contribution is due not to spontaneous fluctuations within the material at a fixed equilibrium temperature, as is the case for brownian noise, but rather, to small fluctuations of the temperature itself. Their effect can manifest in two ways. Firstly, a phase shift in the reflected laser beam is introduced, because the exact position of the mirror surface is moved due to thermal expansion (α) affecting the coating thickness, and thus the optical path *outside of the mirror surface* is altered ("thermoelastic effect"). Secondly, the optical path inside the coating is also modified, both because of thermal expansion α, and because of changes in the refractive index ($\beta = \mathrm{d}n(\mathrm{T})/\mathrm{dT}$) as a function of temperature ("thermorefractive effect"). The coherent sum of these two phenomena is referred to as "thermo-optic effect". To distentangle the various contributions, two separate measurements are needed. In the first place, one can obtain an estimate of the total optical path change inside of the material ($\alpha + \beta/n$) as the temperature varies, with a set-up of broad spectrum light reflected by (or transmitted through) a coating film of the material of interest. The interference between the two coating interfaces (coating/substrate and coating/air) allows the estimation of the optical path, and the procedure can be repeated as the temperature of the sample is made to vary. In second instance, the sole thermoelastic contribution is measured by depositing the same material on a cantilever substrate, with a high contrast in thermal expansion α between substrate and coating, and by measuring the curvature of the sample as a function of temperature, by means of an optical lever. Knowing the total contribution and the thermoelastic contribution (α), the refractive contribution (β) can then be extracted, and the thermo-optical behaviour completely characterised [9].

Finally, it would be remiss not to mention that a notable part of the research effort is also focussed on metrology issues. In the case of mechanical losses, this comprises assessing reproducibility and uniformity of the measuring protocols between laboratories, performing pre-measurement treatments such as lateral laser polishing of the samples, and study of ageing effects. In this respect, a systematic source of error when employing crystalline substrates is the problem of thermoelastic shift. The background noise due to thermoelastic dissipation in the substrate is affected by the presence of a coating, so that simply subtracting the bare sample noise from the coated sample noise, in order to extract the contribution of the coating, is not a viable option in some cases. Efforts have been made to parametrise the thermoelastic shift as a function of temperature, sample geometry and coating properties, in order to quantify it or, alternatively, to find favourable conditions for which it can be considered negligible.

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