Probing the atomic structure of amorphous Ta$_2$O$_5$ mirror coatings for advanced gravitational wave detectors using transmission electron microscopy

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Abstract. Advanced generations of ground-based gravitational wave detectors will use ultra-low-loss amorphous dielectric multilayer mirror coatings in order to minimise thermal noise, a limiting factor in detector sensitivity. Transmission electron microscopy is a promising way to probe the atomic structure of these coatings in an effort to better understand the causes of the observed mechanical loss (internal friction) and hence thermal noise.

1. Introduction

Long-baseline interferometers such as those of the LIGO [1], Virgo [2] and GEO [3] projects are being used in searches for the effects of gravitational radiation emitted from astrophysical sources on the relative displacements of test masses, which are coated to produce highly reflective mirrors. These gravitational wave detectors are the most sensitive displacement instruments ever to be developed and are able to detect strains in mirror displacements at the level of $10^{-19} m/\sqrt{Hz}$, with planned detector upgrades to form ‘Advanced’ detectors aiming at another factor of 10 improvement in sensitivity [4, 5, 6].

Noise from various sources is a major challenge when attempting to reach such sensitivities. At the highest sensitivity range (from a few 10s Hz to several 100 Hz) thermal noise from the test mass mirrors and their suspensions is a significant limitation. The test mass mirrors are made from ultra pure fused silica (SiO$_2$) with multi-layer amorphous dielectric mirror coatings used to provide high reflectivity. Significant levels of thermal noise arise from the mechanical losses of the mirror coatings [7]. The coatings currently used are formed from multiple layers, each of $\lambda/4$ thickness layers of Ta$_2$O$_5$ and SiO$_2$ [7].

The fundamental processes which cause mechanical loss in these coatings are not well understood. The aim of this research is therefore to understand the processes causing the mechanical losses on an atomic scale. Here we use transmission electron microscopy and associated analysis techniques to study, for the first time, the local structures and bonding in argon ion-beam-sputtered Ta$_2$O$_5$ coatings produced by CSIRO [8] which have been heat treated to a variety of temperatures. This is an important preliminary step towards identifying a quantitative relation between atomic structure and macroscopic coating mechanical loss.
2. Experimental methods

The transmission electron microscope (TEM) gives the ability to probe and characterise the atomic structure of the amorphous mirror coatings using reduced density functions (RDFs). Figure 1 (left) shows an image of a typical multilayer mirror coating, the darker sections are the Ta$_2$O$_5$ layers. For the purposes of this research, the samples used for analysis have only a single layer of Ta$_2$O$_5$ deposited on a silica substrate. Figure 1 also shows a typical amorphous diffraction pattern one would get when looking at only the Ta$_2$O$_5$ layer. It is these diffraction patterns that act as the raw data for the RDFs.

![Figure 1. Bright field image of a multilayer coating of dark Ta$_2$O$_5$ and light SiO$_2$ layers (left), CBED amorphous diffraction pattern of Ta$_2$O$_5$ coating (right).](image)

The RDF is a statistical representation of where atoms sit with regards to a central atom [9] and can be effectively described as the Fourier transform of the intensity profile one would get from an amorphous diffraction pattern,

$$G(r) = 4 \int_0^\infty \varphi(q) \sin(qr) dq,$$

where $q = 4\pi \sin \theta / \lambda$, $r$ is the distance from a central atom and $\varphi(q)$ is the reduced intensity function derived from the intensity profile [9]. A detailed guide to the numerical calculation approach for the RDF can be found in the review article by Cockayne [9].

The peaks of this distribution give information about the nearest atomic neighbours such as the peak intensity, which describes the probability of finding a particular atom at a certain distance. Also, thermal vibrations within the atomic structure contribute to the peak widths and may be useful for understanding the measured mechanical loss of the coating which is directly related to the thermal noise.

To gain the maximum amount of data from the RDFs, reverse Monte Carlo modelling can be used to model the amorphous structure. Modelling can also be used to find the interatomic bond angles and co-ordination numbers.

The model is produced by firstly randomly packing a defined number of atoms in a defined area and given the atomic boundary conditions of a typical crystalline phase of the material. It is then compared to the experimental RDF via reverse Monte Carlo simulations where the atoms in the model are re-arranged randomly until they sit in a configuration that agrees with the RDF data, and hence the amorphous phase. Energy optimisation of the model was then carried out using DFT modelling to make sure the atoms are sitting in physically reasonable positions.
3. Results

The RDF and modelling procedure was carried out on three Ta$_2$O$_5$ argon-ion beam sputtered coatings that were heat treated at 300, 400 and 600°C. These coated samples were single layers of Ta$_2$O$_5$ on a SiO$_2$ substrate. The process of heat treatment is to leave the sample in an oven under normal atmospheric conditions at a defined temperature for 24hrs.

The mechanical losses of these samples were measured at cryogenic temperatures primarily as a means of using temperature as a probe to study the behaviour of the internal friction of the material, and also because new generations of detectors may work at cryogenic temperatures to further reduce noise sources. The samples show that higher heat treatment temperatures correspond to an increase in the magnitude of loss peaks observed at cryogenic temperatures [10].

The aim of this experiment was to see if there was any difference in the amorphous structures measured using RDFs.

Figure 2. Average RDFs of coatings heat treated at 300, 400 and 600°C (left), Local structure variation showing a greater difference in RDFs at 300, 400 and 600°C (right).

Figure 2 (left) shows three reduced density functions from a tantala layer of mirror coatings heat treated at 300, 400 and 600°C. The averaged RDFs, where RDFs from various areas in the same sample are averaged, show a negligible change as the heat treatment temperature rises. However preliminary investigations into local structural changes, as opposed to averaged areas, suggest that there may be a slight change in the local ordering as the heat treatment temperature rises as can be seen in the right of figure 2. To explain further, some local areas seem to show better defined RDF peaks in samples that have been heat treated at high temperatures suggesting that, at least locally, the heat treatment may be leading to an increase in local ordering. It is worth pointing out that all the coatings looked at had no signs of crystallisation, therefore any differences measured are changes in short range order as these samples all remain amorphous. Further investigation is required to understand the nature of this change and to relate it to the mechanical loss measurements.

Figure 3 shows the amorphous model generated from the reverse Monte Carlo process and energy optimisation based on the 400°C RDF data. Preliminary results from this model show an average Ta to O bond length of 2.10Å with the presence of short Ta to Ta distances of 3.28Å on average.

What is planned next is to relate this model using the reverse Monte Carlo procedure to the local RDF variation from the 300 and 600°C samples and compare the data, looking for any differences that can be related back to changes in the mechanical loss.
4. Conclusions
We have made major advances in our ability to characterise the atomic structure of amorphous mirror coatings and are working to use this to understand measured mechanical losses. The results from the averaged RDFs of three Ta\(_2\)O\(_5\) coatings heat treated at 300, 400 and 600°C show a negligible change in amorphous structure. However, there may be some local changes from area to area in the samples showing an increase in the Ta to O coordination as the heat treatment temperature increases. Reverse Monte Carlo modelling combined with energy optimization of the amorphous structure provides a promising way to probe the maximum amount of information possible from these samples. The main emphasis of future research will be to understand how these results relate to the mechanical loss of these coatings.

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