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Silicate bonding properties: investigation through thermal conductivity measurements

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Abstract. A direct approach to reduce the thermal noise contribution to the sensitivity limit of a GW interferometric detector is the cryogenic cooling of the mirrors and mirrors suspensions. Future generations of detectors are foreseen to implement this solution. Silicon has been proposed as a candidate material, thanks to its very low intrinsic loss angle at low temperatures and due to its very high thermal conductivity, allowing the heat deposited in the mirrors by high power lasers to be efficiently extracted. To accomplish such a scheme, both mirror masses and suspension elements must be made of silicon, then bonded together forming a quasi-monolithic stage. Elements can be assembled using hydroxide-catalysis silicate bonding, as for silica monolithic joints. The effect of Si to Si bonding on suspension thermal conductance has therefore to be experimentally studied. A measurement of the effect of silicate bonding on thermal conductance carried out on 1 inch thick silicon bonded samples, from room temperature down to 77 K, is reported. In the explored temperature range, the silicate bonding does not seem to affect in a relevant way the sample conductance.

1. Introduction

In designing a cryogenic, high power GW interferometric detector, the selection of materials for the realization of the mirrors and of their suspension stages is a critical item; fused silica masses and monolithic suspensions are no longer suitable, due to a broad dissipation peak in silica around 40 K which spoils its performances [1]. Among the suggested materials, silicon [2] has a very low intrinsic level of dissipation especially at low temperatures ($\phi(300\,\mathrm{K}) = 2.8 \times 10^8$, $\phi(77\,\mathrm{K}) = 5 \times 10^9$ and $\phi(4.2\,\mathrm{K}) = 6 \times 10^{10}$ [3] [4]), large bulk tensile strength (about 7 GPa, surface effects can redduce it to about 200 MPa [5]) and its thermal expansion coefficient α(T) vanishes at about 123 K and 18 K (see figure 1), being almost zero at lower temperatures. Thermal effects coupled with α, like the thermoelastic noise in mirrors and suspension elements, are also zeroed at these temperatures.

Moreover, silicon thermal conductivity is very high. Efficient thermal conduction is needed for the suspension elements to extract the heat deposited within the optics by high power lasers

used to reduce shot noise. In an interferometer operating at cryogenic temperatures, with about 1 MW of circulating power and mirrors with absorption of few ppm, silicon ribbons with a cross section on the order of 100 mm^2 in a monolithic arrangement are suitable for extracting a sufficient amount of heat with just 2 K of temperature difference along the 0.7 m length of the ribbon.

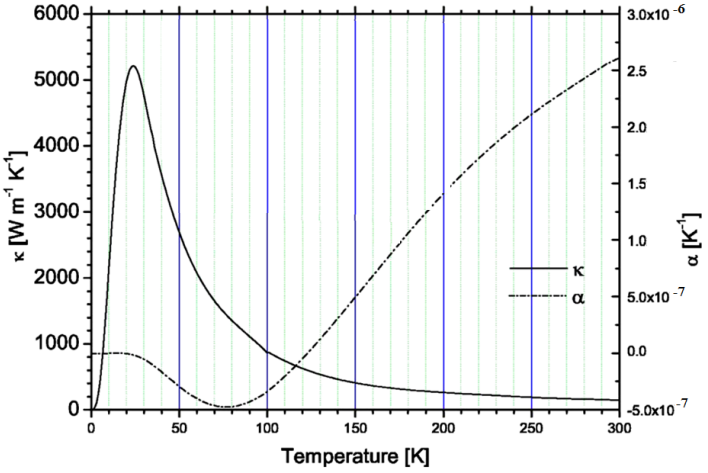

Figure 1. Typical curves of the thermal conductivity $\kappa(T)$ and thermal expansion $\alpha(T)$ for silicon [6] [7].

A typical thermal conductivity curve versus temperature for pure silicon is shown in figure 1. Below 100 K the thermal conductivity κ strongly depends on doping, specimen geometry and crystalline regularity. In particular, in a perfect silicon crystal the peak position is related to the specimen dimensions which limit the mean free path of the phonons, that is dominated by Umklapp scattering at room temperatures. The free path is also sensitive to isotopic mixing: very high thermal conductance has been observed in isotopically pure silicon-28 specimens [8], corresponding to a peak value of $\kappa \simeq 3 \times 10^4$ at 20 K.

In order to realize a monolithic silicon suspension stage, mirror and suspending ribbons have to be joined together. To avoid spoiling the silicon advantages, joints must have high breaking strength and must not introduce intolerably high mechanical dissipation. A highly promising chemistry-based technique for the construction of cryogenic and ultra-low loss monolithic suspensions is silicate bonding. The bonding is realized with a very thin layer of thermal oxide grown on the two silicon surfaces with high flatness (within $\lambda/10$). A drop of aqueous hydroxide solution such as KOH or NaOH is deposited among the surfaces; a silicate gel is formed, that solidifies into a rigid and robust siloxane network. To bond Si samples, surfaces must have SiO_2 coatings about 100 nm thick. The thickness of the obtained bond is on the order of 50 nm, with breaking stresses at room and cryogenic temperatures (77 K) reported to be 39 ± 14 MPa and 39 ± 8 MPa respectively [9].

A complete thermomechanical characterization of the bonding requires the evaluation of its effect on the thermal conductivity across the bonded parts. This feature has to be studied in cryogenic conditions, to check that the heat extraction capability of the suspension is not compromised by it.

2. Measurement technique

In the Firenze INFN Virgo laboratory, we implemented a facility for the measurement of thermal conductivity of materials from 300K down to 4K. The measurement is performed by recording

at each temperature T the regime ΔT set along a sample portion of length L and cross section Σ by a constant heat flux P. The sample is placed in a liquid nitrogen cryostat under vacuum at 10^{-8} mbar, so that heat conduction in air is negligible. Then the conductivity κ is computed as:

$$\kappa(T) = \frac{P}{\Delta T}\frac{L}{\Sigma}$$

For this expression to be valid, the heat flux along the sample must be homogeneous, so that cross sections Σ are isothermal surfaces. To study the effect of the bonding layer, the sample under investigation is built by silicate bonding the flat faces of two silicon cylinders. The bonding of the cylinders was performed in the laboratory of the University of Glasgow using Prolog Semicor, P/Boron doped silicon. To have a situation close to the expected geometry of silicon monolithic suspensions, a cross section of diameter 1" was chosen. We measure the temperature difference both along the entire sample, thus including the bonding contribution, and along one of the two silicon cylinders, to compare with the known silicon curve and to check the goodness of the measurement. The two differences are conveniently obtained by placing three temperature sensors, two on the same cylinder, the last one on the other, beyond the bonding layer.

The presence of damages or inhomogeneities in the bonding layer could perturb the heat flux. To allow the measurement of the overall conductance of the specimen even in such case, the temperature difference must be sensed in places where the flux is unperturbed. Therefore, we performed FEA simulations to draw the isothermal surfaces in case of a small detachment (few mm^2) of the bonding. We found (see figure 2) that at 1 cm from the detachment the flux can be regarded as homogeneous. To place the three sensors at least that distance from contact and bonding surfaces, the bonded cylinders are cut 5 cm and 3 cm long.

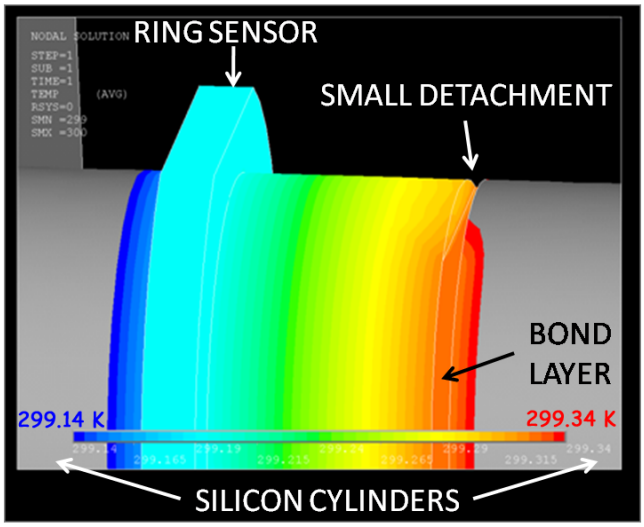

Figure 2. FEA simulation showing in colours the isothermal surfaces when a small detachment in the bond is produced. The ring sensor (see figure 3) is 1 cm from the bonding layer. The bonding layer is indicated by the black arrow. The simulation was performed at room temperature with 1 K temperature difference across the sample.

The experimental setup is shown in figure 3. The constant heat flux is provided by Joule effect in a resistive coil. The flux is made homogeneous by passing through an aluminium heater before reaching the sample. The specimen is placed in contact with the heater and with the

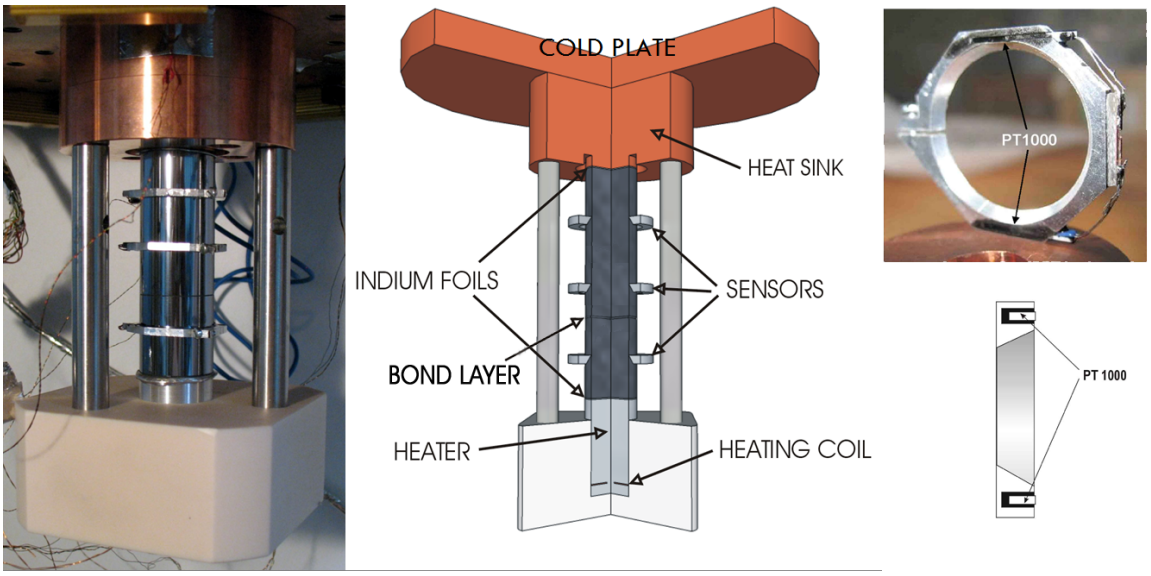

Figure 3. Picture and scheme of the experimental setup described in the text. On the right, a view of an aluminum ring sensor (inner diameter 1") is shown.

heat sink, connected to the cold plate of the cryostat. The quality of the contact is guaranteed by inserting thin indium foils.

Three aluminium rings are placed along the sample, with the inner sharp edge placed in contact with the silicon surface. The temperature of the rings is acquired by PT1000 resistive sensors glued to them. The rings allow a good accuracy in the determination of L.

After the sample was cooled down to 77 K, the measurements were performed during the free rise of the temperature to room value. For each measurement, the collected data enable calculation of the conduction of both a bond-free section of the silicon cylinder and a section that contains a bond.

3. Results and discussion

3.1. Measurement uncertainties

As from the latter equation, the estimated relative uncertainty on the measurement is given by several contributions:

$$\frac{\Delta\kappa}{\kappa} = \frac{\Delta P}{P} + \frac{\Delta L}{L} + \frac{\Delta(\Delta T)}{\Delta T} + \frac{\Delta\Sigma}{\Sigma}$$

The power P is estimated by measuring the voltage and the current in the heating resistive coil. In the typical measurement conditions (P=20 mW), the relative error on P is less than 1%. Due to the very high conductance of the sample, power losses through the coil wires or by radiation are negligible.

The distances L (17.8 mm along pure silicon and 44.8 mm across bonding) have been measured within $\pm$0.3 mm, and the radius of the cross section within $\pm$0.05 mm. Therefore, the contribution to the relative error due to both these measurements is at most 3%.

The temperature difference ΔT is evaluated by measuring the resistance of PT1000 sensors. The resistance is measured in an AC bridge, then converted to a temperature. Therefore, we have:

$$\Delta T = \alpha G \Delta V$$

where α is a conversion factor given by the PT1000 calibration curve, $G = dR/dV$ is the AC bridge gain and ΔV is the bridge voltage. The relative error on α is less than 1%. The bridge

gain was determined by unbalancing the bridge with a known resistance, and its reproducibility was within 1%. The fluctuations of ΔV are very small, so that $\sigma_{\Delta V}/\Delta V \simeq 1\%$ in the worse conditions, that is, at low T where the ΔT is very small (about 4 mK). The setup is sensitive to temperature differences as low as a few tens of μK.

The estimated relative error of the measurement is therefore about 6%.

3.2. Results

The measured values for $\kappa(T)$ are presented in figure 4. The obtained silicon conductivity is compared with pure silicon literature values [10] [7] and with measured values for a P/Boron doped specimen with cross section 6x6 mm^2 found in [11]. The agreement at temperatures not far from room temperature, where the effects of doping and geometry of the specimen are less important, proves that there is no evidence of perturbations due to non-homogeneity of the thermal flux. Also, a curve, computed modeling the bonding layer (plus the oxide layers) as a cylindrical section of glass 300 nm thick, is drawn for comparison.

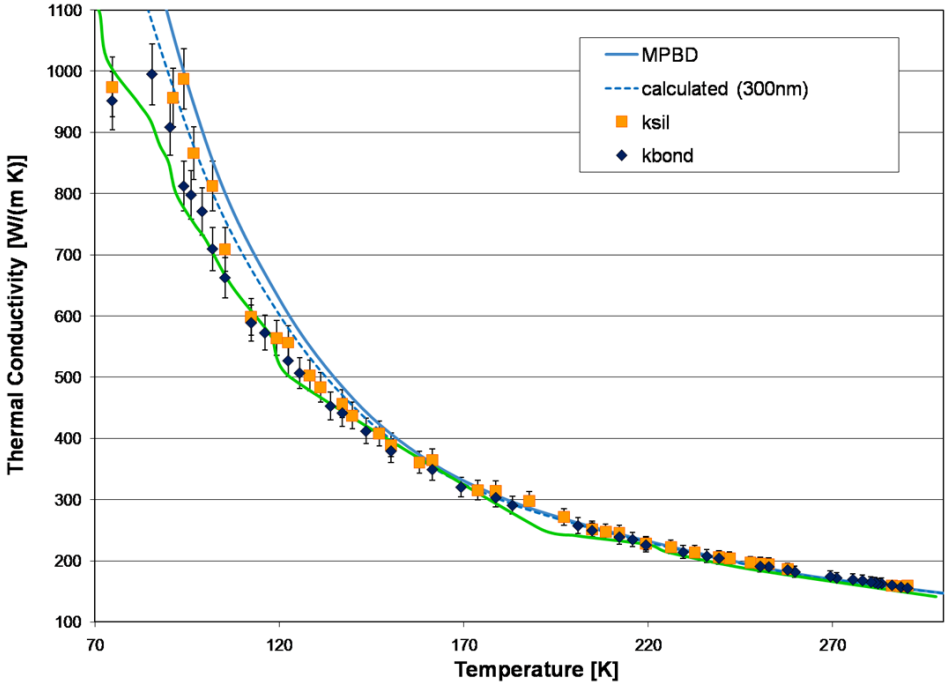

Figure 4. Conductivity measurement results. MPDB is the pure silicon curve from Material Properties Database. "Calculated" is the curve obtained by modeling the bonding as a cylindrical section of glass 300 nm thick. "Ksil" and "Kbond" are the measured values for silicon and across the bonding. For comparison, the green curve is an interpolation of the measured conductivity values for a P/Boron doped specimen with cross section 6x6 mm^2.

In the explored temperature range, the silicate bonding does not seem to affect in a relevant way the sample conductance. In figure 5, the conductivity values measured including the bonding layer, normalized to the measured silicon κ are presented. The effect of the bonding is lower than the measurement error, in agreement with the estimation obtained from the 300 nm glass section model, except for temperatures in the range 77$\div$100 K.

However, measurements from 77 K to 100 K were hard to be obtained in this configuration: due to the rapid rise of the sample temperature in this range, the temperature varied significantly within a measurement, thus making impossible to reach a stationary ΔT. As this error source

cannot be simply estimated, it is not included in the error bars of figure 4. Therefore, in this range of temperatures, our measurements cannot be conclusive.

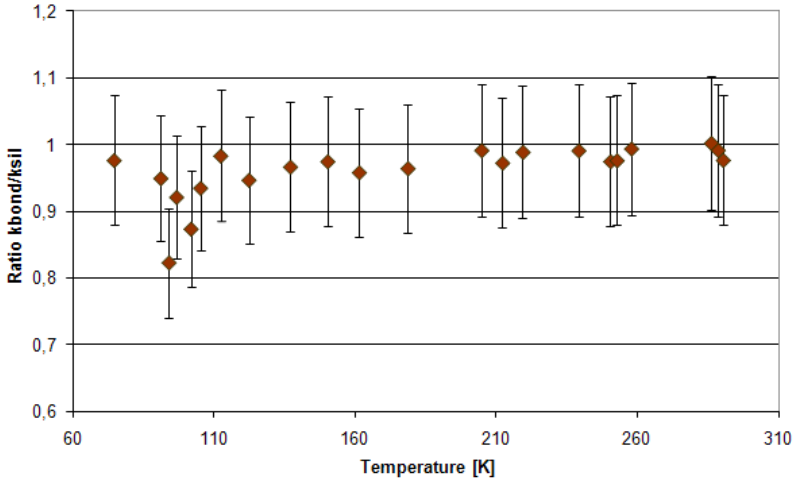

Figure 5. Conductivity values measured including the bonding layer, normalized to the measured silicon conductivity.

The study needs to be extended to lower temperatures in order to fully understand the effect of bonding in cryogenic conditions. Therefore, we plan to carry out the measurements on this sample down to 20 K with liquid Helium and the present setup. After that, new sensing elements and liquid Helium will be used for reaching 4 K. To avoid the problems due to the free rise of the temperature up to 100 K, a temperature control system is going to be implemented in the setup.

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