Realization of a broadband picosecond acoustics setup to study hypersonic propagation in disordered materials.

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Introduction

Glasses and glass transition stand, in much quoted estimate of a Nobel laureate, as "perhaps the deepest and most interesting problem in condensed matter physics"[1].

The classification of solids into crystalline and amorphous is based on the existence of long range order. In a crystal all the atoms are arranged over a three-dimensional periodical lattice, of the same size of the crystal. Glasses, those amorphous solids undergoing a glass transition, lacks instead any long range periodicity.

Glass plays an essential role in science and industry. The optical and physical properties of glass make it suitable for common applications such as: flat glass used for windows, glass doors, transparent walls, windscreen; (these are very useful for modern architectural and automotive application), container glass, such as bottles, jars, drinkware and bowls, glasses are also widely employed in optics, optoelectronic and laboratory equipments, due to their relatively low thermal conductivity, they also find application as thermal insulator (rock wool).

Understanding of glass-former is a very interesting field for both theoretical and experimental physicist; from a theoretical point of view, glasses represent systems where statistical mechanic properties are dominant, and for experimental point of view a comprehension of these system can help for applications such amorphous solids (like plastic) or polymers.

There is no easy explanation and unanimous consensus on the very nature of the glass transition yet, there exist partial theories often providing very different conclusions about the microscopic mechanisms underlying this transition. However, it is universally recognize that some thermodynamics anomalies at low temperature, such as the behaviour of specific heat or thermal conductivity, are related with an excess of states in the vibrational density of states $g(\omega)$ respect to the Debye $g(\omega)$ which describes crystalline solids. This difference in the v-DOS has also important
implications in the acoustic properties. Most specifically, sound attenuation in glasses exhibits non-trivial behaviour over a wide range of frequencies, at variance with the crystalline counterpart where acoustic damping is fully rationalized in terms of anharmonicity and structural defects.

In this thesis we developed an experimental methods to measure acoustic attenuation in the frequency range unaccessible by conventional frequency domain techniques such as Brillouin Light Scattering (≈ 30 GHz) or Inelastic X-ray Scattering (≈ 1 THz). Understanding the propagation of sound waves in this intermediate region (hypersound frequencies) is of fundamental importance for testing lot of theoretical models.

The history of experimental physics goes hand in hand with development of new technologies, and thanks to the development of laser physics now it is possible to have ultrashort light pulses. This paved the way to study sound propagation through timed resolved techniques, i.e. launching an acoustic wavepacket (pump) and probing its fate during its travel inside specimen. Building on recent experimental layout, the so called, interferometric picosecond acoustics (PA), we developed a broadband approach capable to study acoustic properties in a previously unaccessible region with unprecedented sampling efficiency. We demonstrate our setup in the case of vitreous silica solving a long standing controversy on the nature of acoustic damping in this prototypical glass forming system.

In chapter 1 we discuss some basic properties of disordered systems; in chapter 2 we will focus on the behaviour of sound attenuation as determined by spectroscopic techniques in the frequency domain; in chapter 3 we will detail the basics of time resolved approaches picosecond acoustics (PA), used to monitor hypersound propagation in disordered materials; In chapter 4 we discuss a broadband version of PA technique, and the experimental setup that we have realized. Finally, in chapter 5 we present the data analysis for evaluating the sound damping in silica.
Chapter 1

The Glassy State

There are three different phases for a glass-former: crystalline, amorphous and liquid. If the glass-former is in a liquid state, decreasing the temperature we can obtain both crystalline or glassy state. A way to distinguish the two different states is to study the density as a function of the temperature.

In the first case, decreasing temperature, there is a sudden jump of density at the transition temperature know as $T_m$. In the second case the liquid, once reached $T_m$, does not crystallize and becomes under-cooled, i.e. although the crystalline state is more stable, it is still in a liquid state; if one decrease again the temperature, the system reach a temperature $T_g$ where the slope of the density changes gradually and the liquid become a glass. Following this description, it seems that in both cases there is a thermodynamic transition. Actually basing on the Ehrenfest classification, the thermodynamic parameters of first order, such as volume or density, undergo in the two cases respectively a discontinuity of functions (first order transition) and of the derivatives (second order transition); but if one strictly applies this definition, arrives to theoretical prevision (relations about $T_g$ and pressure, thermal conductivity, specific heat) which are not confirmed experimentally. Moreover there are some experimental evidences without an explanation in a purely thermodynamic theory, e.g. in the glass transition $T_g$ depends on the cooling rate. To explain this dependency there was introduced calorimetric theories, where the entropy plays a fundamental role.

For underlining the fundamental differences between crystalline and disordered solids, let us introduce the Debye model.
1.1 The Debye Model

In order to understand the difference between disordered and crystalline materials it is useful to introduce the Debye model, which describes the low temperature properties of crystalline solids. The basic assumption of this model is that the phonon dispersion relation between frequency $\omega$ and wavenumber $k$ is linear

$$\omega = c_D k,$$

where $c_D$ is the Debye velocity. To obtain the right number of phonons it is defined the Debye wavenumber $k_D$ as the bigger phonon wavenumber, i.e. $k \leq k_D$.

Because the total number of phonons for each branch is equivalent to the number of particles in the solid, $N$, we can write

$$N = \frac{V}{(2\pi)^3} \int_{|\mathbf{k}|<k_D} d^3k = \frac{V}{(2\pi)^3} \frac{4\pi}{3} k_D^3,$$

this implies

$$k_D = \left(\frac{3}{6\pi^2} \frac{N}{V}\right)^{1/3}.$$

Figure 1.1: Phase diagram of glass transition.
moreover, it is possible to define the Debye frequency as

\[ \omega_D \equiv c_D k_D \] (1.4)

### 1.1.1 The density of vibrational states

The density of vibrational states for acoustic phonons is the sum of the longitudinal and the two transverse components. First of all we can evaluate the number of phonons with a frequency smaller than \( \omega \)

\[
N_L(\omega) = \frac{V}{(2\pi)^3} \int_{|\vec{k}| < \frac{\omega}{c_L}} d^3k = \frac{V}{(2\pi)^3} \frac{4\pi}{3} \left( \frac{\omega}{c_L} \right)^3
\]

\[
N_T(\omega) = 2 \frac{V}{(2\pi)^3} \frac{4\pi}{3} \left( \frac{\omega}{c_L} \right)^3 .
\] (1.5)

Then the longitudinal and transverse density of vibrational states \( g_L(\omega) \) and \( g_T(\omega) \) can be calculated as

\[
g_L(\omega) = \frac{d}{d\omega} N_L(\omega) = \frac{V}{2\pi^2} \frac{1}{c_T^3} \omega^2
\] (1.6)

\[
g_T(\omega) = 2 \frac{V}{2\pi^2} \frac{1}{c_T^3} \omega^2 .
\] (1.7)

So the total density of vibrational states \( g(\omega) \) is therefore

\[
g(\omega) = g_L(\omega) + g_T(\omega) = \frac{3V}{2\pi^2} \frac{1}{c_D^3} \omega^2
\] (1.8)

with

\[
\frac{3}{c_D} = \left( \frac{1}{c_L^3} + \frac{2}{c_T^3} \right) .
\]

### 1.1.2 The specific heat

Having evaluated the total density of vibrational states it is now possible to calculate the specific heat \( C_V(T) \):

\[
C_V(T) = \frac{d}{dT} \int d\omega \ h_\omega \ n(\omega, T) \ g(\omega)
\] (1.9)
where $n(\omega, T)$ is the Bose occupation number.

This means that:

$$C_V(T) = \int_0^{\omega_D} d\omega \frac{\exp\left(\frac{\hbar \omega}{k_B T}\right) \frac{\hbar^2 \omega^2}{k_B T^2}}{\exp\left(\frac{\hbar \omega}{k_B T} - 1\right)} g(\omega)$$

and in the low temperature limit ($T \ll \frac{\hbar \omega}{k_B}$) this can be written as

$$C_V(T) \approx \int_0^{\hbar \omega_D} d\omega \frac{\exp\left(\frac{\hbar \omega}{k_B T}\right) \frac{\hbar^2 \omega^2}{k_B T^2}}{\exp\left(\frac{\hbar \omega}{k_B T} - 1\right)} g(\omega)$$

where $T_D = \frac{\hbar \omega_D}{k_B}$ is the Debye temperature.

Using (1.3) the (1.11) reads:

$$C_V(T) \approx \int_0^{\infty} d\omega \frac{\exp\left(\frac{\hbar \omega}{k_B T}\right) \frac{\hbar^2 \omega^2}{k_B T^2}}{\exp\left(\frac{\hbar \omega}{k_B T} - 1\right)} g(\omega)$$

$$\approx \frac{3V T^3 k_B^3}{2\pi^2} \frac{T_D^3}{T_D^3} \int_0^{\infty} d\omega \frac{\exp(\omega)}{(\exp(\omega) - 1)^2} \omega^4$$

which is the well known $T^3$ law, describing vibrational component of the specific heat in crystalline materials.

### 1.1.3 Thermal conductivity

The thermal conductivity $\kappa$ due to lattice vibrations in the Debye model can be calculated in a similar way as in the case of an ideal gas:

$$\kappa(T) \approx \int_0^{\omega_d} d\omega \ c_D C_V(\omega, T) l(\omega, T)$$

where $C_V(\omega, T)$ is the spectral component of the specific heat and $l(\omega, T)$ is a scattering length.

For low temperatures $l(\omega, T)$ can be assumed to be constant because it is dominated by scattering at lattice defects and at the surface of the crystal, so it follows that in
the low temperature limit

\[ \kappa(T) \approx T^3. \]  

(1.14)

1.2 Low temperature anomalies in disordered materials

Once we know the results of the Debye model we can compare them with the behaviour of the low temperature properties of the disordered materials. One of the most famous differences is in the behaviour of the specific heat. The Debye model predicts indeed that the specific heat is proportional to the cube of the temperature (1.12). So we expect the ratio of the specific heat over the cube of the temperature to be a constant. Actually this is not the case in the disordered materials where a linear dependency of the specific heat with temperature is found in the low temperature limit, as it is shown in figure (1.2). This can be explained by two-level states [3], which was proposed in 1972 independently by Anderson et al. and Phillips. It is based on the assumption that in amorphous solids some atoms or groups of atoms can occupy two different equilibrium positions. This can be imagined as a double-well potential with respect to some kind of configurational coordinate. The characteristic quantity of this double-well potential is the asymmetric energy, which is the difference of the energies of the two equilibrium positions, and the barrier width and height. The quantum mechanical solution for such a model results in a system with two different energy levels with the distance \( \Delta E \) ("two-level system", TLS) and specific tunnelling probability. In a disordered solid the shape of the double-well potential should vary, which leads to a specific probability density for a certain energy distance \( \Delta E \). If this probability is assumed to be constant, the linear dependency of the specific heat can be explained. Phonons with an energy equivalent to the energy splitting of the two-level system, can be scattered by "the two-level mechanism". With this additional scattering centres, the \( T^2 \) behaviour of thermal conductivity can also be explained. Although this model is quite successful in describing some features of disordered solids, there is no general explanation for the microscopic origin of the two-level system. Moreover, above a few Kelvin, the thermal properties of glasses deviate from the prediction of this model.

Thermal conductivity in disordered solids also shows some anomalies at low
1.2 Low temperature anomalies in disordered materials

Figure 1.2: $\frac{C_V}{T^3}$ of various amorphous solids

temperatures, and as can be seen in figure (1.3), there is an unexpected plateau which is not predicted by the Debye model.

Since both $C_V$ and $\kappa$ are determined by the V-DOS $g(\omega)$, one expects that this latters also deviate from Debye behaviour.

The density of vibrational states, which is shown in figure (1.4), can be measured by means of neutron scattering technique. As it is possible to see there is a deviation from Debye’s law, which predicts quadratic behaviour of the density of vibrational states with frequency. In order to highlight this deviation the $g(\omega)$ is often reported scaled over $\omega^2$ (see figure (1.5)). The resulting peak in figure (1.5) is known in literature as boson peak.

The specific heat of a disordered system can be evaluated via (1.10) using the experimental density of vibrational states. This was done in figure (1.6) which shows that the bump in the specific heat is intimately related to the boson peak.

The interest in physical properties of amorphous solid is not limited to thermodynamic observables, but also acoustic properties such as wave propagation, dispersion relation and the average lifetime of sound waves, measured in recent years through different techniques.
1.2 Low temperature anomalies in disordered materials

Such properties can be measured in the frequency range from $5\,GHz$ to $50\,GHz$, by Brillouin light scattering (BLS) technique. Because of the momentum carried by visible photons, BLS is limited to a range of wavevectors relatively small: $q < 10^{-2}\,nm^{-1}$. For wavenumber larger than $1\,nm^{-1}$ these properties are measured by inelastic X-ray scattering (IXS) technique, a relatively new technique introduced in 1995 [11].

The behaviour of such high frequency acoustic excitations is still poorly understood and it has been studied intensively, both by experimental and theoretical physicist, because of its relation with the boson peak and with the low temperature thermodynamic anomalies.

For example, has been advanced a relation between the boson peak and the existence of localized vibrations; the acoustic wave that became localized could produce the plateau in the thermal conductivity. This theory is based on the assumption that the disorder leads to microscopic random spatial fluctuations of the transverse elastic constant [4]. With this assumption it is possible to demonstrate that the sound

Figure 1.3: $\kappa$ of various amorphous solids
damping ($\Gamma$) is proportional to $\omega^2$ in the whole frequency region covered by the boson peak and it turns to a $\omega^4$ behaviour only below the low frequency edge of the boson peak. Moreover, the frequency dependence of the sound damping is the result of the interplay of several physical mechanisms crucial for many theoretical models. In chapter 2 we will see that 

1) the sound attenuation follows a $\omega^2$ law, according to the Akhiezer mechanism, in the low frequency regime; 

2) there is an intermediate regime, which is hardly accessible by spectroscopic techniques in the frequency domain, where $\Gamma$ follows a $\omega^4$ law and 

3) that in the high frequency regime the sound attenuation follows again a $\omega^2$ law which is due to the structural disorder.

Than in chapter 3 we will discuss in details a pump probe techniques, the picosecond acoustics (PA), which allows to measure the sound damping and the sound velocity in the intermediate regime.
1.2 Low temperature anomalies in disordered materials

Figure 1.4: $g(\omega)$ of glassy glycerol measured by neutron scattering [24].

Figure 1.5: $\frac{g(\omega)}{\omega^2}$ of glycerol.
1.2 Low temperature anomalies in disordered materials

Figure 1.6: $\frac{C_V(T)}{T^3}$ of Glycerol evaluated through the measured density of vibrational states.

Figure 1.7: BLS measure, from this graph it is possible to evaluate the sound velocity and the sound attenuation $\Gamma$. 
1.2 Low temperature anomalies in disordered materials

$q = 1 \text{ nm}^{-1}$

Figure 1.8: example of an IXS measure
Chapter 2

Sound attenuation in disorder materials

Although glasses are disordered materials, both theoretical and experimental physicists have demonstrated, in recent years, the existence of a dispersion relation extending up to wavevectors which are a significant fraction of the first sharp diffraction peak, with pseudo Brillouin zones, defined as the $Q$ region extending up to the first sharp diffraction peak. Then a sound wave in a disordered materials is characterised by a frequency $\nu$ linked to a corresponding wavenumber $Q$.

The sound attenuation and its frequency $\nu = \frac{\omega}{2\pi}$ and wavelength $\lambda = \frac{2\pi}{Q}$ dependence shows a complex behaviour which is the result of the interplay between many competing physical mechanisms, which could be divided into two groups: i) dynamic mechanisms, e.g. the anharmonicity of the interparticle interactions and ii) static mechanisms, e.g. the structural disorder.

In this chapter we will discuss these two contributions and the possible techniques to measure the sound damping in the high and low frequency limits.

2.1 Dynamic mechanism

2.1.1 Theoretical behaviour

The anharmonicity of the interparticle potential (figure (2.1)) rules the attenuation of sound waves in both order and disorder materials. The anharmonic damping of
an acoustic sound wave is characterized by a well defined, temperature-dependent relaxation time $\tau_r$ [5] which defines an high and a low frequency limit for acoustic attenuation.

The anharmonic behaviour is well described by the Akhiezer mechanism, which predicts two different dependency for sound attenuation: in the low frequency limit ($\omega \tau_r < 1 \Rightarrow \omega < \omega_r = \frac{1}{\tau_r}$) this anharmonic contribution dominates the sound absorption [6] and predicts a sound attenuation dependency $\propto \omega^2$. In the opposite, high frequency limit ($\omega > \omega_r$) the sound attenuation coefficient due to this mechanism become frequency independent.

As mentioned there exists a dispersion relation also in glasses; it is therefore possible to define a wavenumber $Q_r$ associated to $\omega_r$:

$$Q_r = \frac{1}{v \tau_r}$$

(2.1)

where $v$ is the sound velocity.

Hence we can say that for wavenumber smaller than $Q_r$ the sound attenuation coefficient due to the anharmonicity of the interparticle interactions scales as $Q^2$ and it become constant for wavenumber larger than $Q_r$.

In the Akhiezer regime a sound wave passing through a solid can be attenuated by two processes [18]. First, if the wave is longitudinal, periodic contractions and dilations in the solid induce a temperature wave via thermal expansion. Energy is dissipated by heat conductions between regions of different temperatures. Second, dissipation occurs as the gas of "vibrons" (the term vibron is refered to any quantized vibrational mode in glass) tries to reach an equilibrium characterized by a local sound-wave-induced strain. The latter is the internal friction mechanism. To establish the relative importance of the two processes, consider these order of magnitude formulas:

$$\Gamma_h \approx \frac{\nu^2}{\rho v_s^3} \frac{\kappa T \alpha^2 \rho^2 v_s^2}{C}$$

$$\Gamma_i \approx \frac{\nu^2}{\rho v_s^3} C T \tau_r \gamma^2$$

(2.2)

Where $\Gamma_h$ is the attenuation coefficient due to heat conductivity, $\Gamma_i$ is the attenuation coefficient due to internal friction processes, $\rho$ is the density, $C$ the specific heat per unit volume and $\alpha$ is the coefficient of thermal expansion. If we introduce the
2.1 Dynamic mechanism

Figure 2.1: The anharmonicity of the potential between particles

Diffusivity $D$ via this formula:

$$\kappa \approx CD,$$

the ratio $\frac{\Gamma_h}{\Gamma_i}$ reads to:

$$\frac{\Gamma_h}{\Gamma_i} \approx \frac{D}{v_s^2\tau_r}.$$  \hspace{1cm} (2.3)

The factor $v_s^2\tau_r$ measures the ability of vibrons to absorb energy from a sound wave of velocity $v_s$.

In crystals $D \approx v_s^2\tau_r$, so the ratio in (2.3) is of order unity; in glasses energy is transferred by diffusion and $D$ is not related to $\tau_r$, and a typical value of that ratio is 0.02, so $\Gamma_h$ can be neglected. Thermal conductivity $\kappa$ is larger in crystals than in glasses, and it is demonstrated that $\tau_r$ strongly depend on the anharmonicity, in particular is smaller in glass materials. After all these considerations and from equation (2.2) one would naively expect the sound attenuation in a glass to be much smaller than in the corresponding crystal, since the anharmonicity leads to a smaller $\kappa$ and $\tau_r$. But $\Gamma$ in glasses is of the same order or even higher than
in crystal; the reason is the internal-strain-induced anomalously large Grüneisen parameters of resonant modes. Resonant modes are low-frequency extended modes whose amplitudes are unusually large at a small, typically undercoordinated, region. The sound attenuation is then also caused by the strong coupling of sound waves and resonant modes; since the glass materials are much more open structures than crystalline solids, have an higher number of resonant modes, and consequently a larger sound attenuation coefficient.

2.1.2 Experimental measurements

The sound attenuation coefficient is proportional to the linewidth $\Gamma$ of the Brillouin peak in the dynamic structure factor $S(Q,\omega)$. Let us focus now on a specific glass former that we have used in our experiments in order to understand the sound damping in glass material: $v-SiO_2$ (silica).

In this system the dynamic mechanisms dominate for wavenumber smaller than $Q_r = 6 \cdot 10^{-2} \text{nm}^{-1}$. Then it is possible to measure the sound damping due to these mechanism through Brillouin light scattering (BLS). Through this technique, indeed, one can measure the dynamic structure factor in the range of small momentum exchange $Q < 10^{-2} \text{nm}^{-1}$. As it is shown in figure (2.2) the $Q^2$ behaviour of $\Gamma$ in the low frequency limit is confirmed by BLS measurements.

Experiments shows also the following features in the low frequency limit: i) At temperatures $T < 10K$ and ultrasonic frequencies $\Gamma(T)$ exhibits a small, frequency-dependent peak. ii) For temperatures between 10 and 200K another peak in $\Gamma(T)$ develops; but it broadens at hypersonic frequencies and is not seen above $100\text{GHz}$. iii) At hypersonic frequencies $\Gamma(T)$ increase slightly with $T$ from temperatures higher than the peak (ii) to at least $300K$. From all these features emerges a temperature dependence of the sound damping in the low frequency limit, especially for ultrasonic waves.
2.2 Static mechanism: the structural disorder

2.2.1 Theoretical behaviour

The low and high frequency limits of the sound attenuation associated with structural disorder are defined by a parameter $Q_R$ which is different in comparison to dynamical mechanism parameter $Q_r$. In fact the typical defect size $\xi = \frac{2\pi}{Q_R}$ plays a fundamental role for the topological disorder.

If the wavelength of the acoustic wave is bigger than $\xi$, the Rayleigh scattering is responsible for the sound damping; this means that if $Q < Q_R$ the sound attenuation coefficient $\Gamma$ scales as $Q^4$. In the high frequency limit ($Q > Q_R$) the Rayleigh scattering regime is abandoned and there are some theoretical models in order to explain the experimental $Q^2$ behaviour of the sound damping in the THz region.

One of them [4] is based on the model assumption that the disorder leads to microscopic random spatial fluctuations of the transverse elastic constant. With this assumption the excess of DOS has been shown to arise from a band of disorder-induced irregular vibrational states. This model provides a first explanation of the plateau of the temperature dependent thermal conductivity and its relation with the boson peak. Moreover it is possible to calculate the shape of the Brillouin resonance and so to evaluate the sound attenuation coefficient $\Gamma$ and this theory shows the $Q^2$ dependence of $\Gamma(Q)$ and predicts a quantitative relationship between the boson peak and $\Gamma(Q)$.

2.2.2 Experimental measurements

Already as mentioned in chapter 1, it is possible to measure the sound attenuation coefficient in the THz region ($Q > 1nm^{-1}$) through inelastic X-ray scattering (IXS). These measurements confirm the $Q^2$ behaviour in the high frequency limit (figure (2.2)) and provide an evidence of existence of a topological disorder mechanism which contributes to sound damping.

In fact it is possible to study, through IXS, the THz excitations in the temperature region where the BLS data in the GHz range show a marked temperature variation. As it is shown in figure (2.3) the $\Gamma(Q)$ measured by IXS is temperature independent in the whole $0.1T_g$ to $T_g$ region, whereas the BLS data increases by a factor 10. These two opposite behaviour provide an important evidence of the existence of at least
two different attenuation mechanism: \(i\) one is dominant at low \(Q\) and is of dynamic origin; \(ii\) the other one is dominant at high \(Q\) and is of static origin.

Consequently, the high \(Q\) behaviour of the sound damping in glasses must have a structural origin, i.e. it must be due to the topological disorder of the glass structure. In figure (2.4) it is reported as an example the disordered structure of the silica.

\[ \text{Figure 2.2: } \Gamma(Q) \text{ from different data sets: Circles, IUVS [19]; triangles and diamonds, visible BLS [9, 11]; solid circle BLS data with 266\,nm photons [10]; squares, IXS [11]. The IUVS measurements show a crossover from low and high frequency limits for } \nu \text{ of about 100}\,GHz. \]

\[ \text{2.3 Sound damping scenario and IUVS technique} \]

The picture coming from the reported data suggests that at least two mechanisms contribute to the sound damping in a glass material: \(i\) dynamical mechanisms such as the anharmonicity of the interparticle interaction and \(ii\) static mechanism...
2.3 Sound damping scenario and IUVS technique

The sound attenuation coefficient can therefore be written as:

$$\Gamma_Q(T) = \Gamma_Q^{(D)}(T) + \Gamma_Q^{(S)}$$

where $\Gamma_Q^{(D)}(T)$ is a temperature dependent dynamic part and $\Gamma_Q^{(S)}$ is due to topological disorder. As aforementioned, at room temperature the dynamical contribution to the sound attenuation scales as $Q^2$ (Akhiezer mechanism) for $Q < Q_r = \frac{1}{\nu T r}$ becoming constant for $Q > Q_r$. Contrarily the statical part scales as $Q^4$ (Rayleigh scattering) for $Q < Q_R = \frac{2\pi}{\xi}$ and scales as $Q^2$ for $Q > Q_R$. But $Q_r$ is different from $Q_R$; so we can summarize the scenario by a three-regime behaviour of the sound damping:

- A low frequency region, for $Q < Q_r$, where the sound damping is dominated by the dynamic processes and $\Gamma(Q) \propto Q^2$.
- An intermediate frequency region, for $Q_r < Q < Q_R$, where the sound damping is dominated by both processes.
2.3 Sound damping scenario and IUVS technique

- An high frequency region, for \( Q > Q_R \), where the sound damping is dominated by the topological disorder.

However, this picture is highly debated because it critically depends on the location of \( Q_r \) and \( Q_R \), and in many glasses they belong in a \( Q \) region which is hardly accessible by spectroscopic techniques in the frequency domain, even if the observation (see figure (2.2)) that the two quadratic behaviours do not have the same coefficient is an important evidence of the existence of the intermediate region. In fact, the IXS data cannot be extrapolated to low \( Q \) values because it would predict width values in excess to the measured \( \Gamma_Q(T) \). This observation excludes that the sound attenuation coefficient scales as \( Q^2 \) in the whole \( 0.01 - 10nm^{-1} \) \( Q \) range.

It is possible to characterize the intermediate region through inelastic UV scattering technique (IUVS) [19]. In fact this technique, using photon energy larger than those of BLS, allows to measure the dynamic structure factor up to a momentum transfer region of \( 0.14nm^{-1} \). This technique has been used to study acoustic properties of silica and it was found a \( Q^4 \) behaviour of \( \Gamma(Q) \) in the crossover region with \( Q_r = 0.11 \pm 0.01nm^{-1} \), as it is shown in figure (2.2). However the silica has electronic absorption in the UV range, so this measurements could be not reliable.
In the next chapter, we will introduce a pump probe technique, the picosecond acoustics (PA), in order to study the hyperson sound propagation in disorder materials and in particular we will focus on the measurement of sound damping in a sample of silica in the intermediate $Q$ region. This is the focus of this thesis, since I have contributed to realization of an experimental setup of a broadband version of PA, for monitoring the hypersonic sound wave propagation in a set of silica samples.

Figure 2.5: In this figure there are the energy-momentum areas covered by different experimental techniques used to monitor sound waves in disordered system. PA technique cover the region between BLS and IXS.
Chapter 3

Picosecond acoustics technique

The Picosecond acoustics technique is an interferometry time-domain method that allows one to accurately measure the attenuation and the velocity of longitudinal sound waves with wavelength in the Brillouin and in the intermediate range. In fact this is a "pump&probe" technique in which an ultra-short optical pump pulse is partially absorbed by a thin metallic transducer layer, deposited onto the surface of the disordered sample. The metallic film acts as transducer launching, by "instantaneous" thermal expansion, a longitudinal acoustic wave-packet with a spectrum in the 40 – 400 GHz range [14]. It is then possible to monitor the propagation of acoustic pulse inside the sample by a delayed optical probe pulse with wavelength $\lambda_{pr}$. The probe pulse is reflected by the strain wave through a stimulated Brillouin scattering process, i.e. interacting with a single phonon frequency:

$$\nu = \frac{v_s Q}{2\pi} = \frac{2nv_s}{\lambda_{pr}} \cos(\theta)$$  \hspace{1cm} (3.1)

where $v_s$ is the sound velocity, $Q$ the acoustic wavevector, $n(\lambda_{pr})$ the refractive index of the medium and $\theta$ the scattering angle. The Brillouin scattered intensity interferes with the portion of beam reflected by the fixed interfaces, leading to time-dependent oscillations with a periodicity $T$ set by the momentum conservation law (3.1):

$$T = \frac{\lambda_{pr}}{2nv_s \cos(\theta)}.$$  \hspace{1cm} (3.2)

Comparing (3.2) and (3.1) one immediately realizes that the time period of
3.1 Generation of phonons

Let us start with the case of a titanium thin film, with a thickness of about 10\,nm, deposited on a substrate of silica (figure (3.2)). When an ultrashort light pulse (the pump) of duration of order 100\,fs is focused on the surface of the titanium, the light energy is "impulsively" absorbed by the electrons in the metal. Because of the high diffusivity of the heated electrons, indeed, the energy is uniformly distributed throughout the film in a very short time ($10^{-14}$\,s). The electrons then relax towards equilibrium transferring their energy to the atomic lattice, i.e. by the emission of thermal phonons in about $10^{-13}$\,s. These phonons ultimately escape into the substrate of silica causing the cooling of the metal. Since the typical time constant for the film to cool off is in the range 100 – 500 ps, for time delays of the order of 100 ps it is possible to approximate the time-dependence of the temperature of the metal as an almost sudden jump $\Delta \Theta$ to a nearly constant value. In view of that the light pump
causes an isotropic thermal stress, $\Sigma$, in the metal given by [13]:

$$\Sigma = -3B\beta \Delta \Theta(r),$$

(3.3)

where $B$ is the bulk modulus, $\beta$ the linear thermal expansion coefficient and $\Delta \Theta(r)$ the sudden jump of temperature.

Within these approximations, and for a Gaussian light beam, it is possible to evaluate $\Delta \Theta$ as [13]:

$$\Delta \Theta(r) = \left(1 - R - T\right)\frac{E}{\pi a^2 C d} e^{-\frac{r^2}{a^2}},$$

(3.4)

where $E$ is the light pulse energy, $R$ the optical reflectivity at the Ti–SiO$_2$ interface, $T$ the transmission coefficient, $C$ the specific heat of the titanium of thickness $d$ and $a$ the width of the pump beam. Moreover, it is reasonable to assume that the motion of the film and substrate is almost entirely in the direction perpendicular to the film surface ($z$ direction) since $a >> d$[13]. Under this condition the thermal stress launches a strain pulse propagating in the forward and backward directions bouncing back and forth between the film interfaces. When these strain pulses are incident onto the Ti–SiO$_2$ interface, they are indeed reflected with a reflection
3.2 Detection of phonons

The motion of the strain pulse into the silica can be monitored using a time-delayed optical probe, as it is shown in figure (3.2). This probe pulse, undergoes indeed two reflections at the interfaces (\(\text{Air} - \text{Ti} \) and \(\text{Ti} - \text{SiO}_2\)) and a weaker reflection at the \textit{moving} strain pulse. The strain pulse produces indeed local changes in the refractive index of the substrate which scatters the probe beam. The three beams interfere constructively or destructively depending on the distance \(v_s t\) travelled by the strain pulse. Then the total reflected intensity is oscillatory in time, according to the Bragg relation (3.2); a measurement of this period can be used to evaluate the sound velocity.

The strain pulse undergoes acoustic damping during its propagation in the substrate. This damping induces a minor local change in the refractive index, and ultimately time dependent attenuation of the reflected intensity. Consequently, the damping rate of the intensity oscillations gives the sound attenuation coefficient.

The PA technique is therefore based on the variation of the reflectivity \(\Delta R\); let us try to evaluate this variation. We will assume, for the sake of simplicity, that the sample is under vacuum condition [15]:

\[
\eta_0 = \frac{1 + \nu}{1 - \nu} \beta \Delta \Theta(r) \frac{2Z_f}{Z_f + Z_s} \frac{v_f}{v_s},
\]

(3.6)

where \(v_{s-f}\) are the sound velocities and \(\nu\) is the Poisson ratio. This strain is obviously longitudinal in nature and represents the \(\eta_{33}\) component of the strain tensor.

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where \(v_{s-f}\) are the sound velocities and \(\nu\) is the Poisson ratio. This strain is obviously longitudinal in nature and represents the \(\eta_{33}\) component of the strain tensor.

\[
r = \frac{Z_s - Z_f}{Z_s + Z_f}
\]

(3.5)

where \(Z_{s-f}\) are the acoustic impedance of substrate and film respectively, a portion of the strain pulse is transmitted into the substrate and it is possible to evaluate the amplitude \(\eta_0\) of the first step of the strain entering into the substrate [13]:

\[
\eta_0 = \frac{1 + \nu}{1 - \nu} \beta \Delta \Theta(r) \frac{2Z_f}{Z_f + Z_s} \frac{v_f}{v_s},
\]

(3.6)

where \(v_{s-f}\) are the sound velocities and \(\nu\) is the Poisson ratio. This strain is obviously longitudinal in nature and represents the \(\eta_{33}\) component of the strain tensor.
3.2 Detection of phonons

strain according to:

\[ \Delta n(z, t) = \frac{\partial n}{\partial \eta_{33}} \eta_{33}(z, t) \]  
(3.7)

\[ \Delta \kappa(z, t) = \frac{\partial \kappa}{\partial \eta_{33}} \eta_{33}(z, t), \]  
(3.8)

A plane wave light field,

\[ E^{(i)} = E_0 e^{i(k_0 z - \omega t)} \]  
(3.9)

impinging at normal incidence on a non-strained sample, generates reflected and transmitted electric fields:

\[ E^{(r)} = r_0 E_0 e^{i(-k_0 z - \omega t)} \]  
(3.10)

\[ E^{(t)} = t_0 E_0 e^{i(k_0 z - \omega t)} \]  
(3.11)

where the reflection and transmission coefficients are:

\[ r_0 = \frac{k_0 - k}{k_0 + k} = \frac{1 - n - i\kappa}{1 + n + i\kappa} \]  
(3.12)

\[ t_0 = \frac{2k_0}{k_0 + k} = \frac{2}{1 + n + i\kappa}. \]  
(3.13)

In order to evaluate the change in reflectivity due to the strain we have to solve the Maxwell’s equations when the optical properties vary with position \( z \):

\[ \frac{\partial^2 E}{\partial z^2} = -\frac{\omega^2}{v^2} (\epsilon + \Delta \epsilon) E(z), \]  
(3.14)

where \( \epsilon \) is the dielectric constant:

\[ \epsilon = (n + i\kappa)^2, \]  
(3.15)

and \( \Delta \epsilon \) is the change induced by the strain.

We consider now the simple case in which the strain is localized at distance \( z' \) from the interface, so we can write:

\[ \Delta \epsilon = F \delta(z - z'). \]  
(3.16)
Then the transmitted wave (3.11) is reflected by this discontinuity and the reflection coefficient can be evaluated solving (3.14):

\[ r_1 = \frac{i k_0^2}{2k} F. \]  

(3.17)

So the total reflected wave is

\[ E^{(r)} = \left(r_0 + t_0 r_1 \tilde{t}_0 e^{2ikz'}\right) E_0 e^{i(-k_0 z - \omega t)}, \]  

(3.18)

where \( \tilde{t}_0 \) is the sample-vacuum transmission coefficient

\[ \tilde{t}_0 = \frac{2k}{k_0 + k}. \]  

(3.19)

We can now easily extend this result for the reflection by a localized strain to an arbitrary perturbation \( \Delta \epsilon(z', t) \), i.e. to an arbitrary strain pulse:

\[ r = r_0 + \frac{ik_0^2}{2k} t_0 \tilde{t}_0 \int_0^\infty dz' e^{2ikz'} \Delta \epsilon(z', t) \]

\[ \equiv r + \Delta r. \]  

(3.20)

The change in reflectivity then reads:

\[ \Delta R = |r_0 + \Delta r|^2 - |r_0|^2, \]  

(3.21)

from (3.15) we obtain

\[ \Delta \epsilon(z, t) = 2(n + i\kappa) \left( \frac{\partial n}{\partial \eta_{33}} + i \frac{\partial \kappa}{\partial \eta_{33}} \right) \eta_{33}(z, t). \]  

(3.22)

Combining these results, the first order change in reflectivity is:

\[ \Delta R(t) = \int_0^\infty dz f(z) \eta_{33}(z, t), \]  

(3.23)
3.2 Detection of phonons

where

\[
f(z) = f_0 \left( \frac{\partial n}{\partial \eta_{33}} \sin \left( \frac{4\pi n z}{\lambda} - \phi \right) + \frac{\partial \kappa}{\eta_{33}} \cos \left( \frac{4\pi n z}{\lambda} - \phi \right) \right) e^{-z/l} \tag{3.24}
\]

\[
\tan(\phi) = \frac{\kappa(n^2 + \kappa^2 + 1)}{n(n^2 + \kappa^2 + 1)},
\]

with \( l = \alpha^{-1} \) the absorption length.

The "sensitivity function" \( f(z) \) determines how the strain at different depths below the surface contributes to the change in reflectivity. The damping of the differential reflectivity is directly related to the energy mean free path \( l \). Hence, we can calculate the sound damping by evaluating the damping of oscillations of differential reflectivity.

Traditionally, PA experiments are implemented using a Ti:sapphire oscillator with a repetition rate of \( \approx 100\text{MHz} \) providing a pump with a wavelength of \( \approx 800\text{nm} \) and a frequency doubled probe with a wavelength of \( \approx 400\text{nm} \), with a duration of \( \approx 100\text{fs} \). In figure (3.3) it is shown a typical spectrum of the oscillatory portion of the differential reflectivity \( \Delta R \) as a function of the time delay. But the frequency of the detected phonon is set by the momentum conservation law (3.1), and for a probe in the visible is limited to a few tens of GHz. In a sample of silica \( n = 1.5 \)
and \( v_s \approx 6000 \text{m/s} \), for instance, the detected frequencies are limited to \( \omega \leq 35 \text{GHz} \) (backscattering condition, \( \Theta = 180 \)), for \( \lambda \approx 535 \text{nm} \).

In the next section, we will describe an improvement of the technique which allows one to reach the hypersound regime, i.e. \( \omega > 100 \text{GHz} \) where acoustic properties of amorphous materials are strongly perturbed by the presence of structural disorder.

### 3.3 Measurement of hypersonic attenuation through PA technique

In the previous section we have seen how it is possible to measure the ultrasonic attenuation in the time domain through the picosecond acoustics technique. However, as we have seen in the second chapter, it is also possible to measure the sound damping in the same frequency range by BLS techniques, i.e. directly in frequency domain. Most of the interesting phenomenology of acoustic properties in amorphous materials, however, occurs over a frequency range which correspond to hypersound waves (\( \omega > 100 \text{GHz} \)).

The frequency of the detected phonons depends on the momentum conservation law (3.1) and Bragg relation (3.2) which relates to the refractive index and sound velocity of the material. Hence, we deposited the sample on a material with much larger refractive index and sound velocity, such as silicon \( (n_{Si} \approx 5.3, v_{Si} = 9360 \frac{\text{m}}{\text{s}}) \), in order to enhance frequency of the detected phonons to the hypersonic regime, following the scheme depicted in fig (3.4).

In this way we detect the phonons in the silicon but differently from frequency domain spectroscopies, where it is possible to detect only thermal phonons, with PA we can monitor a coherent sound wave packet that has travelled into the sample before entering into silicon. By detecting the phonons into the silicon, at the interface with the sample, we can obtain some informations about the sound damping in the sample in a frequency region inaccessible by looking at the light scattered by the sample itself. This can be easily understood as the larger is thickness \( d \) of the sample, the more the acoustic wave will be attenuated travelling into the sample and the smaller will be the initial amplitude of the oscillation detected into the silicon.

By measuring the amplitude of oscillation of the differential reflectivity \( \frac{\Delta R}{R} \) in the silicon at the interface with the sample, for a set of samples with different thickness
of silica $d$, we expect an exponential decay:

$$\frac{A_{Si}}{A_{SiO_2}} \propto e^{-\frac{d}{l}},$$

(3.25)

where $l$ is the energy mean free path of the detected phonon. To account for the amplitude in the silicon $A_{Si}$ is normalized to the amplitude in the silica $A_{SiO_2}$.

Having evaluated the energy mean free path, $l$, we can determine the sound attenuation coefficient $\Gamma$:

$$\Gamma = \frac{\alpha v_{SiO_2}}{2\pi},$$

(3.26)

where $\alpha = l^{-1}$ and $v_{SiO_2}$ is the sound velocity of the silica.

This experimental set-up has been recently demonstrated by A. Devos et al. [17] using a $\approx 800nm$ Ti:sapphire oscillator which provides both the pump and probe (by frequency doubling) and using a diode for detecting the intensity of the reflected beam. A very good signal-to-noise ratio was obtained, with differential reflectivity down to $10^{-7}$, but in a very narrow wavelength range in detection, limited by the oscillator tunability. This results are shown in figure (3.5) and figure (3.6).

![Diagram](image.png)

Figure 3.4: if there is a substrate of silicon there are two possibilities: until a time $t^*$ the strain pulse is detected into the silica (figure on the left), after $t^*$ the strain pulse is detected into the silicon.
3.3 Measurement of hypersonic attenuation through PA technique

Figure 3.5: On the left there are the $\frac{\Delta R}{R}$ oscillations for samples with different thickness $d$, as it is possible to see the period in the silica is bigger than the period in the silicon and in the silicon there is also a strong optical absorption; on the right there is $\frac{A_{Si}}{A_{SiO_2}}$ vs $d$, the expected exponential decay is confirmed.

As is can be observed in figure (3.5), the period of the oscillations in the silica is larger than the period of oscillations in the silicon, by the expected factor $\frac{v_{Si}}{v_{SiO_2}} \frac{n_{Si}}{n_{SiO_2}}$. Although the signal-to-noise ratio is very good (see figure (3.5)), figure (3.6) shows the limited accessible frequency range (5 frequencies) and sampling efficiency of this technique.

We developed a broadband version of the interferometric picosecond acoustics technique, which allows accessing in a single measurement nearly one decade spanning range of the acoustic frequency wavepacket with unprecedented sampling efficiency. In the next chapter we will detail this experimental setup, focusing on the main experimental difficulties that we had to overcome.
3.3 Measurement of hypersonic attenuation through PA technique

Figure 3.6: Full widths $\Gamma$ of the Brillouin peaks evaluated through different spectroscopy techniques; the red squares are the Devon et al. measurements [17]. As it is possible to see, there is no crossover at $\approx 100\text{GHz}$ as in the Masciovecchio measurements. This could be related to the fact that Masciovecchio measurements are in the UV where the silica has electronic absorption (and so this measure are not reliable) or to the fact that Masciovecchio measured the bulk properties while Devos measures the surface properties.

Figure 3.7: PA with a sample of silica on a substrate of silicon; the oscillation are modulated by an exponential decay due to slowly varying, transient changes of the refractive index in the transducer.
Chapter 4

Broadband picosecond acoustics

The main focus of this thesis is the development of a broadband version of the interferometric picosecond acoustics setup described in the previous chapter. The main advantage of a broadband probe is the possibility to phase match (eq. 3.1) in a single measurement all the phonons contained in the acoustic wavepacket launched by the pump pulse, with unprecedented frequency sampling efficiency.

Such a setup is based on 800 nm, 100 fs pulses generated by a 80 MHz oscillator and amplified by a regenerative amplifier. A portion of this beam is used as pump; the remaining energy of the amplified pulses is sufficient to generate white light (visible portion extending from 430 nm to 760 nm), using nonlinear effects, focusing the light pulses in a 2 mm thick sapphire plate, and the white light continuum is used as probe.

The light beam reflected by the sample is dispersed by a diffraction grating onto an optical multichannel analyser (OMA) equipped with fast electronics, allowing single-shot recording of the probe spectrum at the full 1kHz repetition rate of the regenerative amplifier.

In this chapter we will illustrate in details:

- the characteristics of the laser source,
- the optical layout of the experiment,
- how we have been able to acquire a single-shot broadband spectrum with a repetition rate of 1kHz,
- the details of our home-made acquisition software,
4.1 The laser source

Our system is based on 80 MHz oscillator plus a regenerative amplifier with a 1kHz repetition rate.

4.1.1 The oscillator

The seed beam is provided by a Micra oscillator. The Micra is a Ti:sapphire laser oscillator system capable of producing modelocked pulse with a bandwidths exceeding 100nm. The pump pulse for the Micra oscillator is the output beam of the Verdi laser, which provide a 5W beam centred at 532nm. Optimal alignment of the pump beam into the oscillator cavity is maintained by the PowerTrack beam steering system. A feedback loop controls the orientation of the first pump steering optic while monitoring the Micra output power level.

The oscillator itself employs Kerr lens modelocking in combination with an intracavity prism pair to generate low-noise, large-bandwidth and high peak ultrafast pulses. The prisms are adjustable along with a slit assembly to provide tunability of the bandwidth and centre wavelength. The cavity also features automated modelocking initiation by means of solenoid-driven movable mirror mount.

Then the output beam of the Micra (the seed) has ≈ 300 mW of power, a repetition rate of ≈ 80 MHz, a centre wavelength of 800 nm (tunable from 750 nm to 860 nm) and a bandwidth adjustable from 30 nm to over 100 nm.

4.1.2 The amplifier

The regenerative amplifier (RGA) is based on a Ti:Sapphire crystal. This is a commonly used laser crystal, in fact it exhibits high resistance to thermally induced stress, allowing it to be optically pumped at relatively high powers without danger of fracture. Moreover it has an high-stimulated emission cross-section and corresponding high gain.

The optical excitation of the crystal is achieved by pumping with an Evolution-15
4.2 Optical layout of the experiment

In this chapter we describe the setup of our experiment. As we have seen in the previous chapters, the setup is based on a regeneratively amplified Ti:Sapphire laser producing 50 fs, 3.5 – $\mu$J pulses centred at 800 nm with a repetition rate of 1 kHz; a schematic layout of the setup is shown in figure (4.1).

A portion of the laser beam, with an energy up to 5 – $\mu$J, is used for the pump beam which is loosely focused on the sample. The phonons produced by expansion of the titanium are in the frequency range from $\approx 40GHz$ to $\approx 400GHz$, then we used a white light continuum for phase-matching these frequencies. In order to generate this WLC, we focus the other portion of the laser beam in a 2 mm thick sapphire plate, obtaining a broadband continuum with a visible portion extending from 430 nm to 760 nm.

As mentioned, we need to focus pump and probe beams at different time delays on the sample, so we used a delay line for delaying the pump beam. We preferred to delay the pump as WLC generation is a second order process which strongly depends
on the position of the focus on the Sapphire plate; even if the delay stage is properly aligned, any minor mechanical jittering can modify the shape or even the stability of the broadband white spectrum.

Since we aim to expose the detector to alternate adjacent \(\text{pump}_{\text{on}}\) and \(\text{pump}_{\text{off}}\) spectra, as it is shown in (4.1), we used a chopper with 1 kHz frequency. Unfortunately it is not easy to discriminate a priori a \(\text{pump}_{\text{on}}\) and a \(\text{pump}_{\text{off}}\) spectrum, in the next section we will detail how to overcome this difficulty.

As it is shown in figure (4.1), we work in backscattering to maximize the accessible phonon frequencies. The layout only uses reflective optics to avoid a spatial chirp of the beam and a consequent temporal broadening of the pulses.

Differently from classic PA experiments we have to analyse a broadband laser beam, so we need to disperse the reflected beam to the CCD with a monochromator; we use a Princeton Instruments 2500i monochromator with 300 lines, and we report in figure (4.4) the efficiency curve of this grating and the CCD coverage. In figure (4.2) and in figure (4.3) a picture of the delay line and of the monochromator plus CCD are reported, respectively.

Figure 4.1: Schematic layout of the PA setup that we used to measure the sound damping of the silica.
4.2 Optical layout of the experiment

Figure 4.2: Delay line used to delay the pump beam; it is controlled by home-made gui, based on a matlab code.

Figure 4.3: In this picture there are the monochromator and the CCD.
4.3 Single shot detection

For the calculation of $\frac{\Delta R}{R}$ we need to acquire two kind of spectra of the probe: one with the pump focused on the sample and spatially overlapped to the probe pulse ($pump_{on}$) and the other with the probe only ($pump_{off}$); we can write:

$$\frac{\Delta R}{R} = \frac{pump_{on} - pump_{off}}{pump_{off}}.$$  \hspace{1cm} (4.1)

The crucial point is to acquire those two different spectra as close as possible in time in order to minimize unwanted "slow" time fluctuations of the laser. The ideal situation, that we have been able to achieve, is single shot acquisition in alternate exposure of the detector. In this way the only fluctuations are due to the uncorrelated shot-to-shot noise of the laser, which is less than 0.5%.

To achieve shot-to-shot acquisitions we used an optical multichannel analyser (OMA) at the output of a grating monochromator (acton sp 2500i) which disperses the white continuum on a CCD array (the princeton pixis 400, instrument camera). This camera has 1340 horizontal and 400 vertical ($20 \times 20\mu m$) pixels. While we have to use all the horizontal pixels to maximize frequency resolution (every horizontal pixel sees light scattered by a phonon of different frequency), we do not need all the vertical pixels. The ideal acquisition protocol is a loop where every exposure is triggered by the laser and readout in less than 1 ms, repeated at the same frequency of the laser system (1 kHz). More specifically these are all the operations to be performed in a single loop:

1. clear the CCD,
2. wait until a trigger
3. expose the CCD during the scattering event,
4. transfer image array to storage array,
5. readout the CCD.

The time duration of a scattering event is much shorter respect to 1ms being essentially the time duration of the light pulse (50 fs). Since the intensity of
4.4 The acquisition software

We have developed an home made GUI and an acquisition software, based on Matlab platform, in order to record and manipulate the spectra of reflected light. This

Figure 4.4: Efficiency curve of our monochromator; the grating is $1 - \mu$m blaze. If we set the monochromator at 510 nm the CCD cover wavelengths from $\approx 400$ nm to $\approx 600$ nm.

The scattered light is much larger of any background from spurious light, and no mechanical shutter would be fast enough to brake one single pulse, we operate the CCD without a shutter, in continuous exposures between two adiacent readouts.

The readout time depends on the number of vertical line to transfer, so we hard-coded the chip of the CCD reducing the effective area to only 20 vertical lines (enough to accommodate the vertical beam size), in order to minimize the readout time. Despite this reduction, the time duration of a sequence of 1340 pixels is 1.4 ms. We therefore eliminated the fist step of the acquisition sequence (clear the CCD) modifying the DLL library of the CCD, ultimately obtaining a sequence time of 0.7 ms enough to enable a shot-to-shot acquisition. We report in the appendix () the C script that we developed to obtain a function allowing to get a number $N$ of spectra, i.e. repeat $N$ times this acquisition sequence.

4.4 The acquisition software

We have developed an home made GUI and an acquisition software, based on Matlab platform, in order to record and manipulate the spectra of reflected light. This
software allows for finding the zero time delay between pump and probe beams, for optimizing the alignment on the sample and it shows the real time oscillations, for a given $\lambda_{pr}$, of the reflectivity during the acquisition providing a feedback of the experiment. The heart of the software is the C script shows in appendix () which allows to acquire a number $N$ of spectra, adjacent in time, with the CCD. This C script was turned into a Matlab function through the mex compiler, and finally embedded into a software operating in two different modes:

- alignment
- acquisition

### 4.4.1 Alignment

In this operating mode one can set a fixed position for the delay line (hence fixed pump-probe delay) and a number $N$ of spectra to acquire. Since the pump beam goes through the chopper, $\frac{N}{2}$ spectra are of type $pump_{off}$ and $\frac{N}{2}$ are of type $pump_{on}$. As it is shown in appendix () this operating mode calculates:

$$\frac{\Delta R}{R} = \frac{pump_{on} - pump_{off}}{pump_{off}}$$

shot-to-shot and this result is averaged on the $\frac{N}{2}$ different acquisitions.

As it is shown in figure (4.5) this quantity is reported on the left graph for any probe wavelength at given time delay. The entire procedure is repeated in a loop until the user click on stop button, allowing to modify the experimental condition and check in real time the results.

For example, in PA experiments we have to focus the pump and the probe beam on the same area of the sample, but it is very difficult to optimize this overlap of the beams by eyes; with this software we can align the focal lens of the pump maximizing the signal of the reflectivity, shown by the software in real time.

Of course the absolute intensity of the signal, from a given sample, depends on the time delay from pump and probe beam, i.e. from the position of the delay line. All the alignment procedure is done for $\Delta t = 2$ ps
4.4 The acquisition software

Figure 4.5: Matlab acquiring software; on the left there is a graph representing differential reflectivity $\frac{\Delta R}{R}$ as a function of the probe wavelength at fixed time delay. The graph up to the right represents the oscillations of the differential reflectivity as a function of the time delay for a fixed wavelength. The image on the right represents the oscillations as a function of both time delay (x axes) and wavelength (y axes). Both the right graphs are refreshed at every step of the delay line.

4.4.2 Acquiring function

The acquiring mode works similarly to the aligning mode, in fact it determines the shot-to-shot $\frac{\Delta R}{R}$, averaged over a number $N$ of acquisitions set by the user, and it is shown on the left graph. But the delay line is now scabbed from a starting position (corresponding to slightly negative delays) to a final position, with a step chosen by the user. In this mode the software also shows a graph of the oscillations as a function of the time delay for a fixed probe wavelength (set by the user within the CCD coverage), adding a point to the graph for each time step of the delay line (see the graph up to the right in figure(4.5)). A graph down to the right it is shown
4.4.3 How to discriminate the two type of spectra

For acquiring both type of spectra it is convenient to use a chopper running to a frequency equal to half of the repetition rate of the laser, i.e. 500 Hz, triggering both the chopper and the CCD by the laser, the first spectrum (and consequently every odd spectra) can be of type \( p_{\text{pump off}} \) or of type \( p_{\text{pump on}} \) depending on the position of the chopper when one starts the acquisition; There are two possibilities:

- if the chopper is blocking the beam when one starts the acquisition, the first spectrum is of type \( p_{\text{pump off}} \)
- if the chopper is not blocking the beam when one starts the acquisition, the first spectrum is of type \( p_{\text{pump on}} \)

These two possibilities are shown in figure (4.6).

With the setup described so far it is impossible to discriminate \textit{a priori} the two spectra and consequently it is impossible to evaluate the differential reflectivity.

To solve this problem we use the following trick: we generate a trigger for the CCD which starts only when the chopper interrupt the beam, instead of triggering the CCD directly with the laser. It is possible to do it using for instance a National Instruments device card (NIDAQ). This device has some analogical input which
can be used for acquiring the laser trigger, and some analogical output which can generate a new trigger sequence. Moreover, it is possible to trigger the beginning of this new sequence with the chopper. In this way, when one starts the acquisition, the trigger for the CCD is provided from the analogical output of the NIDAQ which starts only when the chopper blocks the pump beam, i.e. the first spectrum and consequently all the odd spectra are of type $p_{\text{pump off}}$ (see figure 4.7).

Having discriminated the $p_{\text{pump off}}$ from the $p_{\text{pump on}}$ it is easy to evaluate the differential reflectivity, as it is shown in appendix () where the Matlab script is reported.

Figure 4.6: This figure represents the two different cases for the timing: the blue line is the trigger of the laser; the green line represent the chopper: when it is up the chopper interrupt the pump and it is possible to acquire a $p_{\text{pump off}}$, when the line is down it is possible to acquire a $p_{\text{pump on}}$; the red and light blue lines represent the two different trigger for the CCD depending on when one starts the acquisitions. In the red the first spectrum is $p_{\text{pump off}}$, in the light blue the first is $p_{\text{pump on}}$.

\section{4.5 The stability software}

The generation of white light continuum is obtained through second order optical processes, so even little changes, like the position of the focus or a variation of the energy of the laser pulse, can cause high instability in the spectrum.
4.6 Application of PA to silica samples

The picosecond acoustics techniques has been applied to a set of samples of pure silica ($\nu - SiO_2$) with different thickness for evaluating the sound damping in the intermediate region (from 120 GHz to 200 GHz).

The samples have been obtained starting from a 0.8 – micron layer grown on
the polished surface of a [111] silicon wafer by oxidation in $N_2/H_2O$ atmosphere in a furnace at 950 °C. The wafer was then cut in five portions, each of them exposed for a different time to an HF-based etching solution to uniformly decrease the $SiO_2$ layer thickness. The resulting thickness values were determined through spectroscopic ellipsometry.

In this way we obtained a series of samples with identical microscopic mechanical properties but different thickness of $SiO_2$. Finally a 10 nm thick Ti transducer layer was deposited onto all samples in a single evaporation process, to ensure identical acoustic wavepacket properties for the whole series.

In the following table are reported the different values of thickness of silica.

We derived the pump directly from the laser (800 nm) and obtained a white light continuum, focusing another portion of the laser beam in a 2 mm-thick sapphire plate, for the probe beam which extends from 430 nm to 760 nm. Then using the aforementioned setup we measured the differential reflectivity as a function of the probe wavelength and of the time delay for each sample, acquiring for about 8-10 hours for each sample (about 10 repetitions).

In the next chapter we will see the data analysis of the measures for extrapolating
Table 4.1: Thickness of the different samples of $SiO_2$ used for evaluating the sound damping in the silica.

the sound damping of the silica in the intermediate frequency region.
Chapter 5

Data analysis

After the acquisition, the data are in the form of a $N_{steps} \times N_{pixels}$ matrix for each repetitions on every silica sample. Where $N_{steps}$ is the number of time-delay steps, and depends on the sample (the larger is the thickness of silica, the larger is $N_{steps}$) and $N_{pixels} = 1340$ is the number of horizontal pixels of our CCD.

An exponential decay modulates to the time-oscillations; this decay is due to slowly varying, transient changes of the refractive index in the transducer. First of all we have to remove this exponential background from all matrices. A possibility is to fit each row with an exponential and then subtract the best fit from the data. However these fits strongly depend on the initialization of the parameters and moreover we should do $N_{samples} \times N_{repetitions} \times N_{pixels} = 5 \times 10 \times 1340$ fits. We used instead an algorithm often adopted in NMR [16] which can remove the exponential decay from an entire matrix, reducing the number of operations to only $N_{samples} \times N_{repetitions} = 50$.

This algorithm works better [16] with noisy data, so first we removed the exponential background from each single matrix, and then we averaged on the different repetitions, in order to obtain only one matrix for each silica sample.

In figure (5.1) we report a graphic representation of 5 matrices corresponding to the different thickness of silica. On the $x$ axes there are the time delays, and on the $y$ axes there are the probe wavelengths. Each probe wavelength correspond to a specific sound frequency, according to Eq. (3.1). Of course the strain pulse is first detected into the silica, and after a time $t^*$, which depends on the thickness $d$ of the silica, the strain is detected into the silicon.

One can clearly see that the period of oscillations in silica is larger than in silicon;
The period of oscillations depends indeed on the inverse of the sound velocity (3.2) and the sound velocity in the silica is smaller than in the silicon:

\[ v_{SiO_2} = 5.95 \frac{Km}{s} \]
\[ v_{Si} = 9.36 \frac{Km}{s}. \]  

(5.1)

For a given probe wavelength the amplitude of the oscillations in silicon immediately after the $SiO_2 - Si$ interface manly depend on [2]:

1. How much the corresponding Fourier component of the acoustic wavepacket has been attenuated while travelling through the $SiO_2$ film;

2. how efficiently the acoustic wavepacket reflects the probe pulse in silicon: in fact smaller is the probe wavelength, bigger is the amplitude, this is due to the enhancement of the $Si$ acousto-optic coupling upon approaching the direct band gap value at 364 nm.

We have repeated the measurements for a set of samples with different thickness of $SiO_2$ for getting rid of the second dependency; So we expect an exponential decay of the amplitudes in the silicon close to the $SiO_2 - Si$ interface.

In figure (5.3) is reported the oscillations in two sample, with 230 nm and 313 nm of silica, for a fixed probe wavelength ($\lambda_{pr} = 468$ nm). There is no appreciable damping in the silica, because the mean free path of the phonons in the silica is much bigger than the thickness $d$ of the silica; on the other hand there is a strong optical damping in the silicon.

Obviously the figure (5.3) represents a row of the third image in figure (5.1), we are interested in the values of the first oscillation in the silicon for each row in each matrix, weighted by the intensity in the silica to account for possible changes of the overall signal intensity when switching from one sample to another of different thickness. In the next section we will explain the model for sinusoidal fit used for evaluating the two different oscillation.
Figure 5.1: Differential reflectivity $\frac{\Delta R}{R}$ as a function of time (x axes) and probe wavelength (y axes) for 5 samples of silica with different thickness (230nm, 313nm, 483nm, 642nm, 811nm) on a silicon substrate.
Figure 5.2: Reconstruction of $\frac{\Delta R}{R}$ from the fit.
5.1 Fit model

In order to evaluate the two different amplitudes (in the silica $A_{SiO_2}$ and the first in the silicon $A_{Si}$) we need a model for fitting the data. Under the following assumptions:

- the oscillations are both sinusoidal;
- the oscillations in the silica last until a time $t^* = \frac{d}{v_{SiO_2}}$;
- there is no appreciable damping in the silica;
- the oscillations in the silicon begin after $t^*$;
- there is optical damping in the silicon;
- a fraction $R$ of the strain pulse is reflected by the $SiO_2 - Si$ interface,

we use the following model for fitting each row of each matrix:

$$
Model = A_{SiO_2} \cos\left(\frac{n_{SiO_2}(t - \phi_1)}{\lambda}\right) \left[1 - \Theta\left(t - \frac{d}{v_{SiO_2}}\right)\right] + R \cdot A_{SiO_2} \cos\left(\frac{n_{SiO_2}(t - \phi_1 + d\phi_1)}{\lambda}\right) + A_{Si} e^{-\frac{(t - \frac{d}{v_{SiO_2}})}{\tau_0}} \cos\left(\frac{n_{Si}(t - \phi_2)}{\lambda}\right) \Theta\left(t - \frac{d}{v_{SiO_2}}\right)
$$

(5.2)

Where $n$ is the refractive index in the two different material, $\Theta$ is the Heaviside function, $v$ is the sound velocity, $\lambda$ is the probe wavelength and $\Gamma_0$ is the optical damping.

We have expressed the dependency of the cosine as a function of the probe wavelength $\lambda$, instead of the sound frequency $\nu$, through the Eq. (3.1); in this way we have reduced the number of fit’s parameters to only 7: the two amplitudes, the two phases $\Phi_1$ and $\Phi_2$, the thickness $d$ of $SiO_2$, the probe wavelength and the fraction of sound wavepacket reflected by the $SiO_2 - Si$ interface. We used the tabulated values for sound velocity, refractive index and silicon optical damping.

For each sample of silica we have fitted each row with an algorithm in a loop, so we initialize the fit’s parameter only for the first row, and use the final parameter values of the fit for initializing the following fit.
5.1 Fit model

In figures (5.3) and (5.4) are reported some fits and as it is possible to see the model (5.2) describes properly our data. In figure (5.2) we reconstruct the oscillations of $\frac{\Delta R}{R}$ from the fit.

Figure 5.3: Differential reflectivity oscillations as a function of time delay for a fixed probe wavelength. Until $t^*$ the light is scattered by phonons propagating into the sample, after $t^*$ from phonons propagating into the silicon. The period of oscillation in silica is bigger than in silicon because both sound velocity and refractive index are bigger in silicon. Moreover in silicon there is optical absorption. The thicknesses of silica are respectively: 230 nm and 313 nm
Figure 5.4: Differential reflectivity as a function of the time delay for a fixed probe wavelength. The thicknesses of silica are respectively 483 nm, 642 nm and 811 nm.
5.2 Evaluating sound damping

Once we have evaluated the first amplitude of the oscillations in the silicon we can evaluate the sound damping in the silica; in fact as aforementioned:

\[ \frac{A_{Si}}{A_{SiO_2}} \propto e^{-\frac{l}{l}}. \]  \hspace{1cm} (5.3)

Where \( l \) is the phonons mean free path which is linked to the sound attenuation:

\[ \Gamma = \frac{v_{SiO_2}}{2\pi l}, \]  \hspace{1cm} (5.4)

where \( \Gamma \) is the sound attenuation coefficient which corresponds to the full width at half maximum of the Brillouin peak in a BLS experiment.

So we have evaluated \( l \) as it is shown in figure (5.5).

Through the Bragg relation we have obtained that our measures provide the sound attenuation coefficient for sound frequency \( \nu \) in the range from \( \approx 100 \text{GHz} \) to \( \approx 200 \text{GHz} \).

The quality of the exponential fit varies significantly with the frequency \( \nu \) and for frequencies below 150GHz deviations are observed. In fact the analysis of the interference pattern in this experimental configuration is intrinsically more complex than a simple metal–SiO\(_2\) layout. For example one has to consider also the reflection of the probe from the SiO\(_2\)–Si interface, which can cause Fabry Perot effects [2]. However it seems that these other reflections are relevant only for probe wavelength bigger than 150GHz [2], this is confirmed also from the Root Mean Squared Error (RMSE) which is larger for frequencies smaller than 150GHZ.

Finally we evaluated the sound attenuation coefficient as a function of the sound frequency in the intermediate region, as it is shown in figure (5.6). The broadband PA data clearly indicate a crossover at \( \nu \approx 175 \text{GHz} \), which represent the frequency associated with \( Q_r \), where the sound attenuation goes from low frequency limit of Akhiezer mechanism (\( \Gamma \propto \nu^2 \)) to the low frequency limit of the structural disorder mechanism where \( \Gamma \) is expected to scale at least as \( \nu^4 \) (Rayleigh scattering). So the picture described in chapter 2 for the sound damping behaviour seems to be confirmed, even if the frequency crossover value is larger than Masciovecchio’s result.

In order to emphasize this result, we report in picture (5.7) the sound attenuation
coefficient $\Gamma$ obtained: in this work with broadband PA, with BLS in visible region, with BLS in UV region and with not broadband PA.

![Figure 5.5: Fit for evaluating the sound damping. For frequencies larger than 150 GHz the exponential decay (5.3) is confirmed; for frequencies smaller than 150 GHz the exponential thickness-dependent behaviour of the intensity is modulated, probably by Fabry Perot effects [2].](image-url)
5.2 Evaluating sound damping

Figure 5.6: Sound attenuation coefficient $\Gamma$ measured through broadband PA (red dots) and Root Mean Squared Error (blue line). There is clearly a crossover at $\nu \approx 175 \text{ GHz}$

Figure 5.7: Sound attenuation coefficient measured through different methods.
Conclusions

In this thesis we developed an experimental method (broadband PA) to measure acoustic attenuation in a wavelength and frequency range unaccessible by conventional frequency domain techniques such as BLS or IXS, with unprecedented sampling efficiency. While PA is a pump&probe technique developed in recent years and based on a single frequency detection, we implemented a broadband version of this method based on the generation of white light continuum probe pulses by means of an ultrafast amplified source. We tested this method on a set of Silica samples, with different thicknesses, grew on a Silicon substrate. This experiment showed the existence of a crossover in the sound attenuation behaviour at \( \nu \approx 175 \) GHz, which implies the transition between two different attenuation mechanism of sound waves into disordered materials: dynamical, in the low frequency limit (i.e. anharmonicity of interparticle potential) and static, in the high frequency limit (i.e. structural disorder). This result reconciles the previous measures done by Masciovecchio [19] (BLS) and Rufflè [17](traditional PA), because it demonstrates the existence of a crossover and its dependence on sample details (bulk-oxidized-CVD).

In the first part of this thesis we have emphasized the link between the hypersonic waves propagation in a glassy materials and the thermodynamic low temperature anomalies, such as the specific heat that in a glass does not scales as \( T^3 \) which is the behaviour in a crystalline solids; or thermal conductivity which exhibits a plateau.

In the second chapter we set the theoretical scenario for the description of the sound damping in disordered materials, which can be summarized as follow: in the low frequency limit, for \( \nu < \nu_r = \frac{1}{2\pi\tau_r} \) where \( \tau_r \) is a temperature dependent relaxation time, the sound damping is dominated by dynamic processes, like the anharmonicity of the interparticle potential and the Brillouin linewidth \( \Gamma \propto \nu^2 \); there is an high frequency region, \( \nu > \nu_R = \frac{v_s}{\xi} \) where \( v_s \) is sound velocity and \( \xi \) is the
Conclusions

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typical defect size, where the sound damping is dominated by static process like structural disorder and $\Gamma \propto \nu^2$ but with an higher coefficient; since that $\nu_r \neq \nu_R$, there is an intermediate region where sound damping is due to both processes and is expected to scale at least as $\nu^4$ (Rayleigh scattering). The low and high limits are confirmed by spectroscopy measurements in the frequency domain, such as Brillouin Light Scattering (BLS) (in the low limit) and Inelastic X-ray Scattering (IXS) (in the high limit); but all these technique leave the intermediate region unexplored.

In the third chapter we focused our attention on a specific glass-former: $v - SiO_2$ (vitreous silica), and we detail a pump&probe spectroscopy technique, resolved in the time domain, which allows one to monitor hypersonic sound waves propagation into disordered systems in the intermediate frequency region. BLS is a “fluctuation” experiment, i.e. we can monitor the existing thermal phonons, whereas PA allows one to study a coherent propagating phonons wave packet induced in the sample: an ultrashort light pulse ($\approx 100 fs$) is focused on a transducer metal thin film which adsorbs the light energy and emits a phonons wavepacket in the substrate of silica. These phonons are monitored by a time-delayed light probe, which undergoes some static reflections (by air – metal and metal – SiO$_2$ interfaces) and a dynamic reflection by phonon wavepacket; all the reflected beams interfere constructively or destructively depending on the position into the sample of the propagating sound wave. by measuring the differential reflectivity of the sample, we detect some oscillations, and from that oscillations we can evaluate the sound velocity and the sound damping of the silica.

In chapter 4 we introduced our realization of a broadband version of PA which allows one to monitor hypersound propagation in disordered system with unprecedented sampling efficiency, and present the experimental setup used for applying broadband PA to a set of silica sample.

Finally in chapter 5 we analysed data in order to find sound attenuation coefficient of silica in the intermediate frequency region, $100GHz < \nu < 200GHz$, and we found a sound attenuation scenario with a crossover at $\nu \approx 175GHz$, bridging the low frequency region where $\Gamma \propto \nu^2$ to the intermediate frequency region where $\Gamma \propto \nu^4$. 
Appendix A

Article
A
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Appendix B

Matlab scripts

```c
#include<stdio.h>
#include<stdlib.h>
#include"master.h"
#include"pvcam.h"
#include"mex.h"
#include"matrix.h"

void AcquireSTandard(int n acquisizioni, uns16 frame[]) {
    rgn_type region = { 0, 1339, 1, 0, 19, 28 };

    /* camera handle */
    char cam_name[CAM_NAME_LEN];
    uns32 size;
    //uns16 *frame;
    int16 status;
    int16 hCam;
    /* camera handle */
    uns32 not_needed;
    int16 numberframes = 100;
    int i;
    uns32 skip on = true;

    /*inizializzazione cam */
    pl_pvcam_init();
    pl_cam_get_name( 0, cam_name );
    pl_cam_open(cam_name , &hCam, OPEN EXCLUSIVE );

    pl_set_param( hCam, PARAM_SKIP_SREG_CLEAN, 1 ); //prova
    //pl_set_param( hCam, param id skip serial clear, &skip on ); //riga di prova
    /* Init a sequence set the region, exposure mode and exposure time */
```
pl_exp_init_seq();
pl_exp_setup_seq(0, n_acquisizioni, 1, &region, STROBED_MODE, 10, &size);

//frame = (uns16*)malloc( size );

/* Start the acquisition */
/* ACQUISITION LOOP */
//for(i=0;i<numberframes;i++) {
  numberframes--;
  pl_exp_start_seq(0, Frame);
  /* wait for data or error */
  while( pl_exp_check_status( 0, &status, &not_needed ) &
       (status != REAOUT_COMPLETE & status != REAOUT_FAILED) );
/* Finish the sequence */
pl_exp_finish_seq(0, frame, 0);
/* Uninit the sequence */
pl_exp_uninit_seq();
//free( Frame );
}


void mxFunction( int nlhs, mxArray *plhs[], int nrhs, const mxArray *prhs[] ) {
  //int16 *x;
  uns16 *y;
  int dim[3];
  double *inp;
  int i;
  inp = mxGetPr(prhs[0]);
  x = (int)inp[0];
  dim[0]=1;
  dim[1]=148/*x*/;
  //double appoggio[90000];
  //appoggio[0] = mxCreateDoubleMatrix(1, 90000, mxREAL);
  //plhs[8] = mxCreateDoubleMatrix(1, 90000, mxREAL); /* Create matrix for the return argument */
  plhs[8] = mxCreateNumericArray(2, dim, mxUINT16_CLASS,mxREAL);
  /* Assign pointers to each input and output. */
  y = mxGetPr(plhs[8]);
  AcquireStandard(x,y);
}
flag_stop1;
while(flag_stop2)

STATUS = PV.CancelClose(0); % chiude la cas
STATUS = PV.CancelOpen(0); % apre loc

PV.CAMset(0, PARAM.SHTR.OPEN MODE, .4); % sempre aperto se gia' aperto.
% default=1 apre prima di ogni frame
PV.CAMset(0, PARAM.LOGIC OUTPUT, 1); % non scan 1=shutter
PV.CAMset(0, PARAM.EXF.REINDEX, 0); % 1=reset i tempi in microsecondi
PV.CAMset(0, PARAM.SHTR.CLOSE_DELAY, 1); % varie il tempo di chiusura dello shutter 0=always open
PV.CAMset(0, PARAM_CUSTOM_CHIP, 1); %abilita il custom chip
PV.CAMset(0, PARAM_CUSTOM_TIMING, 1); %abilita il custo timing
PV.CAMset(0, PARAM_3AR_SIZE, 1); % riduce il numero di righe del chip, e quindi il readout
PV.CAMset(0, PARAM_3AR_SHIFT_TIME, 15200); % riduce il tempo di readout, default=15200, minimo 3200
PV.CAMset(0, PARAM_READOUT_PORT, 1); % default=4 setta la port 3bit-Capac,Low-noise
PV.CAMset(0, PARAM_SPOTAB_INDEX, 0); % setta il PIXTIME, 1-500 (VELCCE, 29us), 0=10000 (lenti)
PV.CAMset(0, PARAM_GAIN_INDEX, 1); % default=lowest gain, 3=highest gain
PV.CAMset(0, PARAM_3R_SIZE, 1); % riduce la finestra spettrale, utile in
% appross per ridurre il readout
PV.CAMset(0, PARAM_3RAR_Cycles, 0); % numero di cicli di cleaning, default=1
PV.CAMset(0, PARAM_3MM_OF_3STRIPS_PER_3CLR, 1); % default e' il numero di linee, pertanto non sebra ridurre il readout
PV.CAMset(0, PARAM_3LEAR_MODE, 0); % default=low cleaning, se non riesce a
% recupera, e' readonly
PV.CAMset(0, PARAM_PREMASK, 0); % default=1, riduce la filettatura, non
% loose bene che fa meglio non toccarlo
PV.CAMset(0, PARAM_POSTMASK, 0); % default=1, riduce la filettatura, non
% loose bene che fa meglio non toccarlo
PV.CAMset(0, PARAM_PRESCAN, 0);
PV.CAMset(0, PARAM_POSTSCA, 0);
PV.CAMset(0, PARAM_SKIP_AT_ONCE_BLK, 0); % default=1, riduce 0.7misecondi, il default pero' e' 5
PV.CAMset(0, PARAM/custom_RECEIVE, 0); % default=1, riduce 0.7misecondi, il default pero' e' 5
PV.CAMset(0, PARAM/PIPE/LATCH, 0); % default=1, riduce 0.7misecondi, il default pero' e' 5
PV.CAMset(0, PARAM/PIPE/SPREAD, 0); % default=1, riduce 0.7misecondi, il default pero' e' 5
% acquisizioni (le prime 20 vengono buttate)
% flag_stop=1;

daqreset

ao = analogoutput('rtda', 'Dev1');
addchannel(ao, 1);
ao.SamplingRate = 20000;
ao.TriggerType = 'HwDigital';
ao.HwDigitalTriggerSource = 'PFI1';
putsamples(ao, 0);

if check_background==1
    set(handles.flag_background, 'BackgroundColor', [1 0 0]); % led diventa rosso
    pause(1);
    DATA = acq_roi(1000, 20);
    n_col = int32((length(DATA)/1340));
    DATA = DATA; % reshape per poter usar reshape
    DATA = double(DATA); % trasforma in float
    B = reshape(DATA, 1340, n_col); % reshape multidimensionale su n acquisizioni
    background = sum(sum(B(40:end, 20:end)))/((size(B,1)-40)*(size(B,2)-20));
    clear DATA n_col B;
    set(handles.flag_background, 'BackgroundColor', [0 1 0]); % led diventa rosso
else

    if from==to
        Controller.MOV(axisname, from);
        while (Controller.IsMoving(axisname))
            pause(0.01);
        end
        position = Controller.cMOV(axisname);
    set(handles.pos_del_line, 'String', num2str(position));

end

end
putdata(var1, trigger(1:(n_acq+1)*20:10000));
start1(42)
DATA=sqy_roi(n_acq,20); %acquisisce il primo parametro il numero di acquisizioni, il secondo il numero di pixel verticali della roi
wait(as, n_acq,2);

n_col=int16((length(DATA)/1940));
DATA=DATA'; strafigura per poter usare reshape
DATA=double(DATA); %la trasforma in float
reshape(DATA,1940,n_col)-background; %reshape multidimensionale su n acquisizioni

nkip=20;
even=2;in_col=1;
odd=1;in_col=1;
C=8;even./B(:,odd);
binno=suc(C(:,nskip:length(odd)),2),./size(B(:,nskip:length(odd)),2);
F=mux(B(:,even(nskip:length(odd))),2),./size(B(:,even(nskip:length(odd))),2);
Poff=mux(B(:,odd(nskip:length(odd))),2),./size(B(:,odd(nskip:length(odd))),2);

aves(handles.gain)
plot(lambda1,binno);
aves(handles.aves)
plot(lambda1,F,lambda1,Poff)
data_binned=var1(1940,4);
data_binned(1:end,1)=lambda1(1:end);
data_binned(1:end,2)=binno(1:end);
data_binned(1:end,3)=F(1:end);
%
fr(check_save=2)
string='average.dat';
filenames_average=[path_string];
save(filenames_average, 'data_binned', '-ASCII');
elseif(check_save==3)

    string1='average.dat';
    %string2=num2str(j);
    %string3='.dat';
    string4='all_bin.bin';
    %string5='.bin';
    filename_average=[path string1];
    filename_all_bin=[path string4];
    save(filename_average,'data_binned','-ASCII');
    save(filename_all_bin,'lambda',DATA','-mat');
end

else
    if(from>=to)
        pass-=pass;
    end
for i=from:pass:to
    if(flag_stop)
        Controller.MOV(axisname,i);
        while(Controller.IsMoving(axisname))
            pause(0.01);
        end
        position=Controller.qMOV(axisname);
        set(handles.pos_del_line,'String',num2str(position));
        putdata(ao,trigger(1:(n_acq+1)*20+10000));
        start(ao)
        DATA-acq_roi(n_acq,20);
        wait(ao,n_acq+2);
data.binned=zeros(1340,4);
data.binned(1:end,1)=lambda(1:end);
data.binned(1:end,4)=Cbinned(1:end);
data.binned(1:end,2)=Pon(1:end);
data.binned(1:end,3)=Poff(1:end);
%
if(check_save==2)
    string1='average_DL,'
    string2=num2str(position);
    string3='.'dat'
    path=[path string1 string2 string3];
    save(path,'data.binned','-ASCII');
elseif(check_save==3)
    string1='average_DL,'
    string2=num2str(position);
    string3='.'dat'
    string4='all_bin_DL,'
    string5='.'bin'
    path=[path string1 string2 string3];
    save(path,'data.binned','-ASCII');
    save(path,'lambda','DATA','-mat');
end
else
    break
end
Bibliography


