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New pathways to control the evolution of the atomic motion in metallic glasses

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Abstract

Metallic glass formers are a relatively new entry in glass physics, which has attracted large interest in both physics and materials science communities due to the unique mechanical and structural properties of these materials. Physical aging is however one of the main obstacle to their widespread use as it affects their properties at all length scales. The knowledge of the microscopic mechanisms inducing aging and relaxation is therefore extremely important for both fundamental and applied sciences. In this article we present a review of the recent advances made with the x-ray photon correlation spectroscopy technique on the study of the collective particle motion and physical aging in metallic glasses at the atomic level. We show that a careful tuning of the sample preparation or the application of specific thermal protocols have the potential to drive the glass into more aged or rejuvenated microscopic configurations with different stabilities.

Introduction

Glasses are essential materials in our daily life and they have accompanied progresses in civilization for millennia. Although they are used in a wide range of applications, spanning from engineering to medicine or environmental science, a full comprehension of the mechanisms responsible for their unique properties is still missing [1]. Within the broad family of glassy materials, metallic glasses (MGs) are considered a relatively young entry. Discovered in the nineteen sixties, in few decades they have obtained a dominant role in material science and are currently considered among the most actively studied metallic materials [2]. MGs present an outstanding combination of properties such as high strength, large elastic elongation and high corrosion resistance. In addition, in the temperature region between the glass transition temperature and the crystallization temperature, MGs can be easily deformed in very precise

complex shapes, which makes them promising candidates for many small-scale devices for advanced technological applications, like fuel cell electrodes, storage devices and micro electro-mechanical systems [3]. Metallic glasses are extremely interesting materials also for fundamental science. Thanks to the nature of the metallic bonds, their microscopic dynamics is subject only to translational motion, which make them model candidates for the study of the glass transition nature. MGs share also many similarities with colloidal materials [4], and can be used to find common properties between hard and soft glass formers materials.

As all glasses, MGs are unstable with time and exhibit physical aging. This means that all their properties evolve spontaneously with time and are strongly influenced by the material production and processing. Physical aging can be particularly severe in MGs, more than in other glasses like network or molecular systems, and can lead to ductile to brittle transitions, fractures and crystallization. Aging can therefore produce annoying technological drawbacks, since it makes the amorphous arrested state difficult to control during technological applications [5], [6]. In this respect, the knowledge of the microscopic mechanisms responsible for relaxation and aging in MGs is critical as it allows to predict and control the behaviour of glasses over time and during applications.

In the last decade, coherent x-rays and in particular the x-ray photon correlation spectroscopy (XPCS) technique have emerged as a powerful tool to probe the collective atomic motion of MG during aging [7], [8]. This technique monitors the fluctuations in the scattered intensity generated by the impinging of coherent x-rays on disordered systems, and provides information on the intermediate scattering function (ISF), f(q,t), and thus on the electron density fluctuations at a length scale $2\pi/q$ defined by the probed wave-vector q [9].

Aging studies on MGs have revealed a complex behavior [7]. At the microscopic level, aging is intermittent and dominated by the interplay between density-driven rearrangements associated to the release of residual stresses, and ordering processes at length scales corresponding to the medium range order, which do not affect the local density [10]–[12]. Numerical simulations and elastic measurements suggest avalanche-like mechanisms triggering the relaxation in glasses [13]. Aging is found hierarchical even at the macroscopic scale, and multiple relaxations have been reported during long time enthalpy recovery and stress relaxation measurements in polymers and metallic systems [14]–[16].

After briefly recalling these results, we present here some recent attempts which have been made to control the evolution of the collective particle motion and therefore the aging in MGs

by applying external parameters such as stresses, thermal cycling or specific sample preparations.

Atomic motion across the glass transition

Metallic glasses are usually produced by rapidly cooling the corresponding melt with rates of the order of 10^4-10^6 K/s. Such a fast quenching is fundamental to avoid crystallization and freeze the system in an amorphous solid state. On approaching the glass transition temperature, T_g, from the liquid phase, the collective particle motion evolves from diffusive at high temperatures to a regime of activated dynamics in the supercooled liquid phase [17], [18]. In this region the intermediate scattering function splits in two main processes: i) a fast microscopic decay associated to the rattling of the particles in the cage formed by the neighbouring particles, and ii) the main structural relaxation process corresponding to the escape of the particles from their cages [19]. The latter process corresponds to the long-time decay of the ISF and can be modelled by the empirical Kohlrausch-Williams-Watts (KWW) function $f(q,t)=f_q\exp(-t/\tau)^{\beta}$. In this expression, τ is the structural relaxation time and describes the time necessary for the system to reorganize the structure in a more stable configuration at the probed length scale. In the liquid phase, $\tau(q)$ is equal to the equilibration time and is proportional to the viscous flow. The parameter $\beta(q)$ provides instead information on the degree of heterogeneity of the dynamics and on the nature of particle motion, and $f_q(q)$ is the non-ergodicity parameter representing the long-time plateau preceding the structural relaxation.

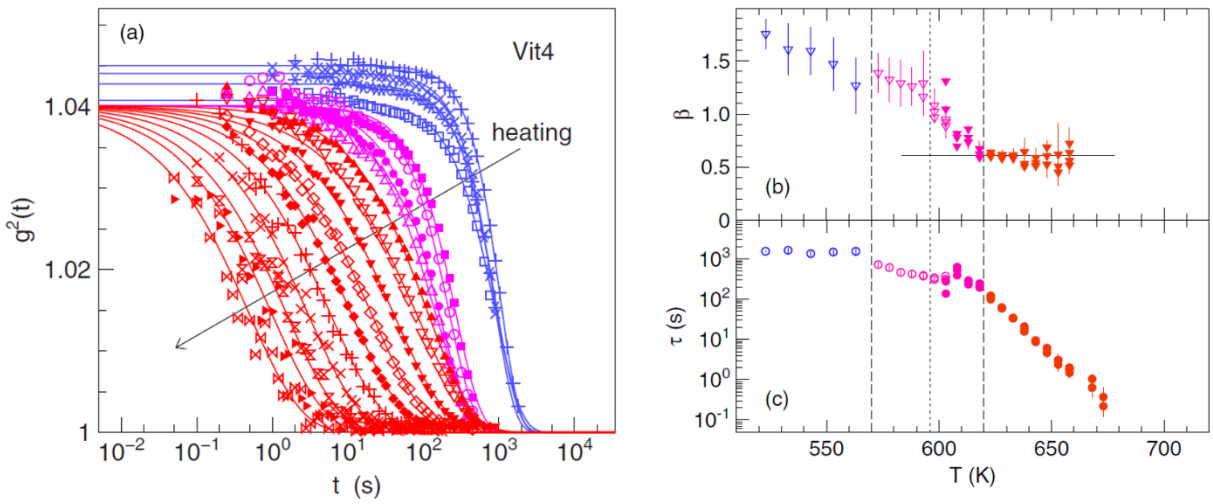

Figure 1. a) Temperature dependence of the correlation functions measured with XPCS in a Vit4 MG during isothermal steps of 5 K between 520K and 675K, obtained by heating the glass with a rate of 1 K/min. The as-cast sample was annealed at 600K prior to the measurements to remove eventual stresses related to the sample preparation. The data are collected at q=2.6Å^{-1} corresponding to the first maximum of the atomic structure

factor. b) and c) Corresponding shape parameter (b) and structural relaxation time (c) obtained by modelling the data in a) with a KWW function. Taken from [20].

Upon further decreasing the temperature, the structural relaxation time increases by several orders of magnitude until the material solidifies in a metastable glassy state. Studies on MGs have shown that the glass transition is accompanied by a dynamical crossover at the atomic level where the decay of the ISF evolves from the well-known stretched exponential shape (i.e. described by $\beta<1$) of supercooled liquids, to an unusual compressed form in the glass which can be modelled with an exponent $\beta>1$ [4]. The same transition is reported also by heating from the glassy state. An example of the dynamical crossover occurring in MGs at the glass transition temperature is reported in Fig. 1a. This figure shows the temperature evolution of the intensity-intensity correlation functions, $g_2(q,t)$ measured with XPCS at the atomic scale upon heating a glass of Vit4 from ambient temperature up to the liquid phase with a heating rate of 1 K/min [20]. In this case, the as-cast glass was relaxed at 600K prior to the measurements to remove the presence of stresses related to the fast-quenching used during the sample preparation. XPCS measurements were performed at the maximum of the static structure factor during isotherms every 5K from 520K up to 675 K. We recall that the $g_2(q,t)$ functions are proportional to the intermediate scattering functions being $g_2(q,t)=1+f(q,t)^2$ [9]. The corresponding KWW parameters τ and β are shown in Fig. 1b and 1c. Three different dynamical regions have been reported for MGs and are visible in the figure. At low temperatures (blue data), the correlation functions vary little with temperature, with an almost constant structural relaxation time and a constant shape parameter $\beta>1$, which has been attributed to the occurrence of a stress-induced dynamics [20]. In the glass transition region (pink data), τ increases in an Arrhenius way with a small activation energy, and is accompanied by a continuous decrease of β toward a stretched value $\beta<1$ which is then reached for temperatures, *T*, larger than the glass transition temperature T_g=600 K. At these temperatures, the system is in the liquid phase and the relaxation time decreases with the typical super Arrhenius behaviour of supercooled metallic liquids (red values) and is accompanied by a constant stretched parameter describing the decay of the correlation functions, close to the value reported with mechanical measurements [20]. Several attempts have been made to explain the compressed decorrelation in the glassy state, from both theoretical and numerical sides [21], [22]. A comparison with macroscopic mechanical measurements associates the dynamical crossover to the separation between the anelastic and viscoelastic response to mechanical stresses at T_g, the former being active also well below T_g

[23]. Numerical simulations in metallic liquids glass-formers report on the existence of compressed intermediate scattering functions in the high temperature liquid phase which appear related to an increasing number of icosahedral clusters on approaching the vitrification and generate additional frustration in the material [24]. Whether a similar argument could be applied to the glassy state is not verified yet. Experiments in soft glasses report instead on a very similar dynamical crossover in a variety of systems including colloids, foams, biopolymers and microgels measured with both x-ray and light scattering photon correlation spectroscopy [25], [26]. In these cases, the crossover is often accompanied by a transition from diffusive dynamics, i.e. described by a sub-quadratic dependence of the relaxation time, $\tau(q)=1/q^2$, to an anomalous super-diffusive, ballistic-like particle motion with $\tau(q)\approx 1/q$. Stress relaxation in the disordered structure can qualitatively describe this phenomenology, although a detailed understanding is still missing [21], [22], [27]. At present, the idea is that the dynamics of soft glasses are triggered by random collapses of the structure due to highly stressed regions in the material. In MGs these regions could be associated to the presence of elastic inhomogeneities at the nanometric scale [28], which could be related to the presence of flow units [2]. Information on the wave-vector dependence of the dynamics in MGs could help clarifying the nature of the dynamical crossover. Due to the out-of-equilibrium nature of glasses, such information is very difficult to obtain below T_g both experimentally and with numerical simulations and only few information about the dynamics in the supercooled liquid phase have been reported so far [18].

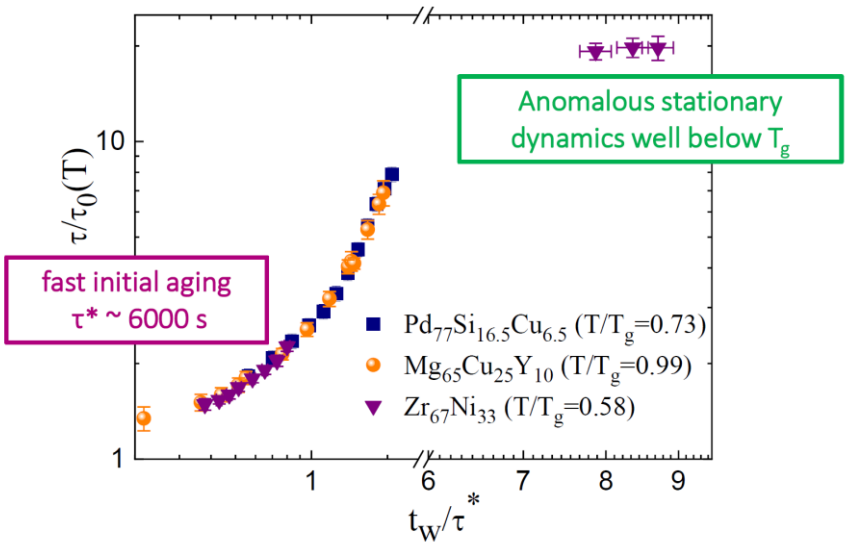

Figure 2. Temporal evolution of the structural relaxation time measured with XPCS in binary and ternary rapidly quenched MG obtained upon heating the as-cast glasses up to specific temperatures in the glassy state with slow heating rates (3K/min for $Pd_{77}Si_{16.5}Cu_{6.5}$ and $Zr_{67}Ni_{33}$, 1K/min for $Mg_{65}Cu_{25}Y_{10}$). The data are rescaled for the initial value of the relaxation time at each temperature, $\tau_0(T)$ and reported as a function of the time elapsed since

the beginning of the isotherm (waiting time, t_w) rescaled by a parameter τ^* that describes the short time exponential growth rate during aging though the expression $\tau(T, t_w) = \tau_0(T)\exp\left({t_w}/{\tau^*}\right)$. $Pd_{77}Si_{16.5}Cu_{6.5}$ data are taken from Ref. [11], $Mg_{65}Cu_{25}Y_{10}$ data from Ref. [4] and the $Zr_{67}Ni_{33}$ data from Ref. [29].

The main difficulty arises from the impossibility to compare data acquired at different times due to the strong evolution of the microscopic dynamics due to physical aging. The first studies on the dependence of the collective atomic motion on aging have been performed on rapidly quenched MGs, i.e. on ribbons of amorphous metals which were produced with a fast cooling rates of 10^6 K/s from the melt and then subsequently heated up in the glassy state. Studies on binary and ternary fast quenched MGs, showed the occurrence of two dynamical regimes which are summarized in Fig. 2: a first exponential increase of the relaxation time during isotherms in the glass, followed by an anomalous stationary regime at longer annealing times (see Fig. 2). X-ray diffraction studies suggest that the first fast dynamical regime arises from density release mechanisms likely involving the annihilation of defects or excess free volume stored in the system, while the stationary regime would be more related to ordering processes occurring at a length scale corresponding to the medium range order [11]. During this stationary regime, the system is trapped in a metastable state from which aging seems then to pursuit through irregular, intermittent evolutions of the collective particle motion which would bring the system in another metastable state. An example of this intermittent dynamics is shown in Fig. 3 for a multicomponent ribbon of $Au_{49}Cu_{26.9}Si_{16.3}Ag_{5.5}Pd_{2.3}$ measured during an isothermal annealing of 5 h at 377 K, well below the glass transition temperature T_g=396K of this composition. The figure reports the temporal evolution of the two-times correlation functions (TTCF), another important quantity which can be extracted from an XPCS experiment. The TTCF is a time resolved version of the standard intensity-intensity correlation functions and provides information on the temporal evolution of the correlation between configurations measured at different times t_1 and t_2. Each pixel on the TTCF expresses the correlation between the intensity of two frames acquired at times t_1 and t_2 in a colour coded fashion. The main diagonal from bottom left corner to the top high corner indicates increasing elapsed times from the beginning of the measurement and corresponds to the condition t_1=t_2, hence showing maximum correlation in red. Horizontal or vertical lines (in the lower and upper quadrants respectively) from a single point along the main diagonal corresponds to correlation measured at increasingly longer delay time $\Delta t=t_2-t_1$ from this fixed reference frame. The loss of correlation (blue values) occurs when the system has relaxed in a new configuration. The width of the yellow profile

along the main diagonal is therefore proportional to the structural relaxation time, the larger this width is, the slower is the dynamics.

In the specific case of Fig. 3a, the TTCF shows two stationary regimes, termed (a) and (b), with constant intensity profiles along the main diagonal but different relaxation times, being the dynamics in the regime 'b' slower than in 'a'. A short temporal period of aging leads to a 'smooth' transition between these two distinct metastable configurations after about 7000s from the beginning of the isotherm. Standard intensity-intensity correlation functions can be obtained by temporal averaging the frames corresponding to each stationary regime ((a) and (b)) and can be used to quantify the increase in the relaxation time induced by the aging, as shown in Fig.3c.

Similar intermittent aging dynamic has been reported in few other compositions and often dominates the dynamics when the glass is subjected to mechanical deformations [30]. Although there is a common idea of an avalanche like particle motions triggering the aging, a clear explanation for the observed behaviour is still missing. Multiple aging regimes have been observed in few macroscopic studies in metallic, polymeric and chalcogenide glass formers [14]–[16]. In these works, the authors report on the existence of multiple decays during enthalpy relaxation interconnected by stationary regimes suggesting a strong connection with the atomic scale dynamics.

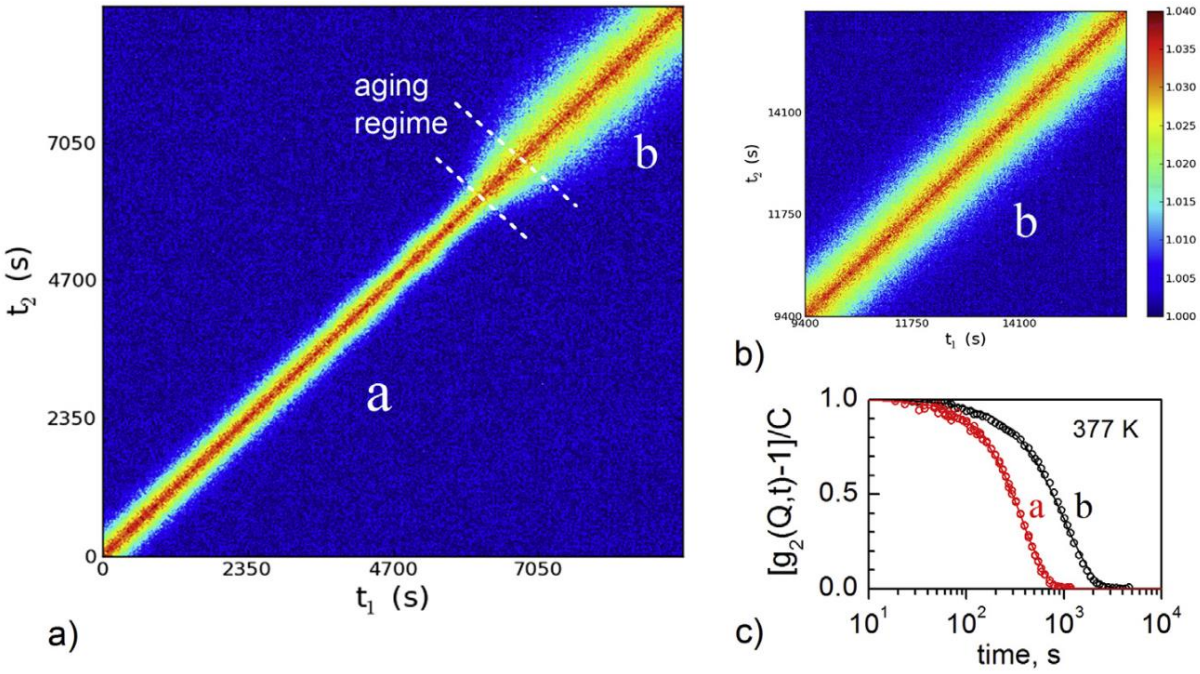

Figure 3. a) TTCF measured with XPCS in an Au-based alloy during isothermal annealing at 377 K for 5 h below T_g. Data show a crossover between two stationary regimes, termed *a* and *b*, connected by a short period of aging occurring after about 7000s from temperature equilibration. b) Continuation of the TTCF at longer annealing times

showing that the dynamics keep stationary in the regime *b* for longer times. c) Intensity correlation functions calculated by temporal averaging the data collected in the two stationary regimes reported in panel (a). Taken from [15].

Controlling the aging through external parameters

Last decades have witnessed an impressive amount of excellent scientific works aimed toward the control of the relaxation behavior and physical aging in MGs by using physical vapor deposition techniques [31], or by altering the relaxation state by applying external parameters, such as stress [32] or extreme thermal cycling [33].

Since the excellent works of Ediger and his group on organic glassy compounds [31], the possibility to dramatically improve the stability of a glass by creating the amorphous solid state by means of physical vapour deposition techniques has fascinated the glass community. The basic idea is that during the deposition the particle mobility at the free surface is orders of magnitude larger than in the bulk and allows for a more efficient particle packing, lowering the energy of the system and bringing therefore the glass in a deeper minimum of the potential energy landscape. Ultrastable glasses formed by physical vapour deposition are considered as an analogue of glassy materials aged for geological times. Among their different physical chemical properties, their improved thermal stability, signalled by a higher calorimetric glass transition temperature on heating, makes them excellent promising candidates for many technological applications. The first vapour deposited MG was produced by the group of K. Samwer and exhibited an increased stability of about 10K with respect to the standard fast-cooled glass [34]. This work paved the way to a large number of studies, although the possibility to obtain metallic ultrastable glasses has been largely debated [35]–[37]. Structural studies of vapour deposited MG have shown that by lowering the deposition rate is possible to improve the stability up to 60K with respect to standard cooled glasses (Fig. 4 a). Curiously, this improved stability comes with a less ordered structure (Fig. 4 b) with respect to an ordinary rapidly cooled MG [38].

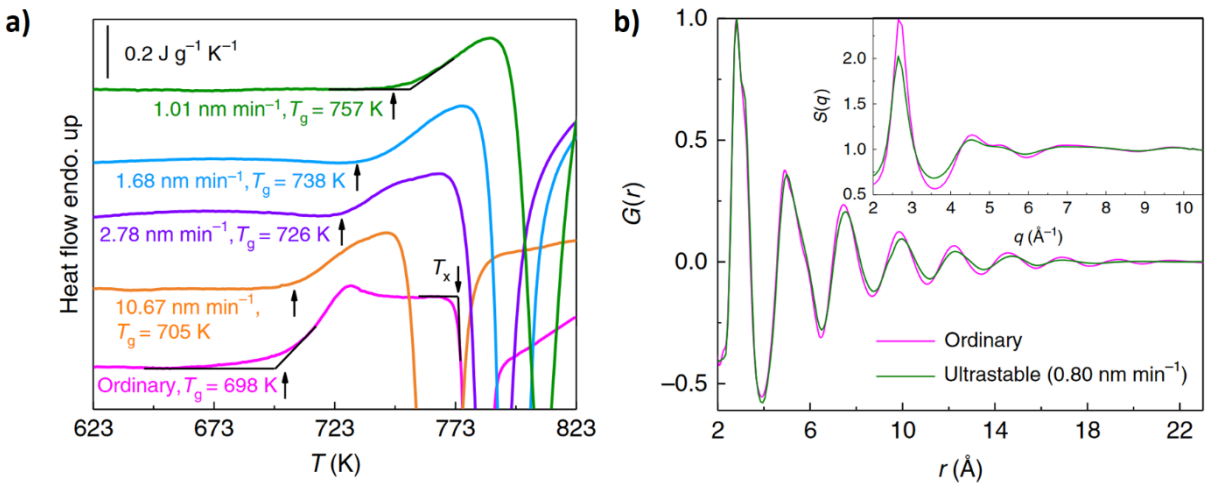

Figure 4. a) Differential scanning calorimetric profiles of an ordinary (i.e. fast quenched) MG and MGs produced by physical vapour deposition highlighting the increasingly stability and thus larger T_g on lowering the deposition rate. An increase of T_g up to 60 K can be obtained with the slowest deposition rate. b) Reciprocal space (inset) and real space static profile of an ordinary and a vapour deposited MG showing larger stability in the calorimetric profile. The improved stability comes with a less ordered structure than in a standard MG. Taken from Ref. [38].

The influence of the ultrastability on the collective particle motion and aging has been investigated in an MG of $Cu_{50}Zr_{50}$ produced by physical vapour deposition following the procedure reported in Ref. 34 [35]. Signatures of an improved stability have been reported at the atomic scale in vapour-deposited MG. This is shown in Fig. 5a which reports correlation functions measured with XPCS in different vapour-deposited MGs: as-prepared, and pre-annealed at different annealing temperatures T_a, above and below the calorimetric T_g and then cooled back to ambient temperature prior to the measurements. Data collected during an isotherm in the glassy state show that the relaxation dynamics is slower in the as-cast vapour deposited MG with respect to that measured on the same material, pre-annealed at higher temperatures (Fig. 5). This means that the highest stability, and thus the slower relaxation time, is obtained with the vapour deposition protocol, while it decreases with further thermal annealing. This process, called in the work *anti-aging*, is opposite to the standard slowing down of the relaxation dynamics due to physical aging induced by thermal treatments. Indeed, in conventional MG, the dynamics slows down significantly by pre-annealing the desired glass just below its T_g [7]. This process does not apply to vapour deposited MG as their structure already corresponds to the most stable state and thus to a deeper minimum configuration in the potential energy landscape.

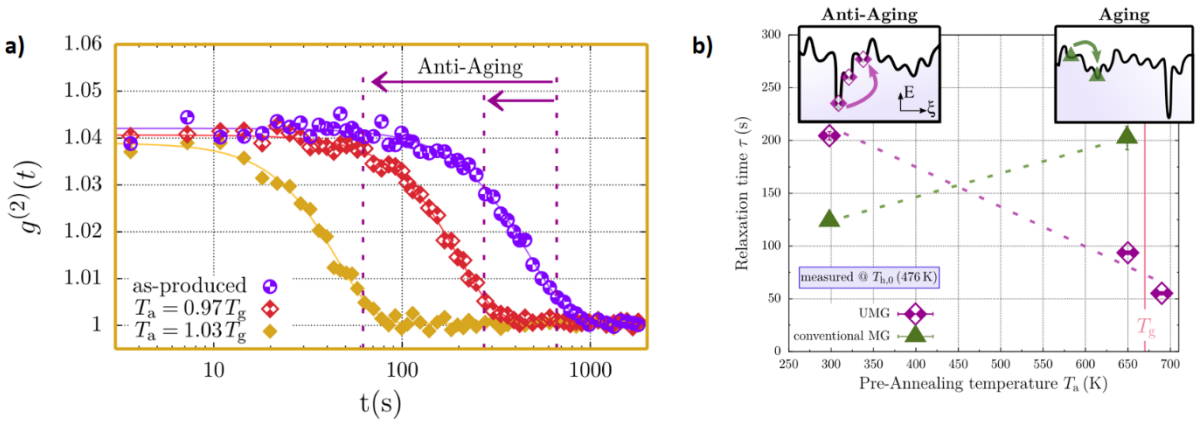

Figure 5. a) Intensity scattering functions measured with XPCS in a $Cu_{50}Zr_{50}$ ultrastable glass as-cast and pre-annealed at different temperatures T_a (1 min annealing, heating rate: 7K/min). Data have been acquired after heating the samples in the glassy state at 476 K. The increased stability of the vapour deposited glass is signalled by the longer decay times (violet data) with respect to the same glass annealed at higher temperatures prior to the measurements. b) Relaxation times corresponding to the data in the left panel (violet diamonds) together with relaxation times measured in a standard fast-quenched MG which was pre-annealing at different temperatures T_a prior to the experiment (1 min annealing, heating rate: 7K/min) and then measured with XPCS at 476K. While the pre-annealing at high temperature slows down the dynamics of the standard MG due to aging, the ultrastable glass loses its improved stability with thermal heating. Taken from [35].

Similar anti-aging has been observed in metallic glasses recovered from anelastic and plastic deformation. The applied shear promotes the glass to a higher energy state evidenced through exothermic events upon further annealing [39], with reported values including a sevenfold increase of the relaxation enthalpy in a $Zr_{50}Cu_{40}Al_{10}$ MG between the annealed and deformed states [40]. Deformation also moves the glass structure toward an apparent lower density and higher disorder state [41], [42], in an opposite way with respect to the aging described above.

Shear stress can also alter the microscopic dynamics as it triggers plasticity localized in narrow shear bands, which accommodate the stress and eventually lead to catastrophic failure. Structurally, these bands are characterized by high disorder, density changes [43], [44], nano-cavities [45], and potential nano-crystals [46], [47]. Typical dimensions of x-ray beams are much larger than the thickness of the shear bands, 5-10 µm against 10-200 nm respectively [45], [48], which prevents the monitoring of the dynamics within the shear bands with XPCS. However, results from site-specific XPCS in the vicinity of a single shear band in a $Zr_{65}C_{25}uAl_{10}$ MG revealed an acceleration of the atomic scale dynamics 10 µm away from the shear band (Fig. 6a), a length scale greater than the shear band classical thickness by three orders of magnitude [49].

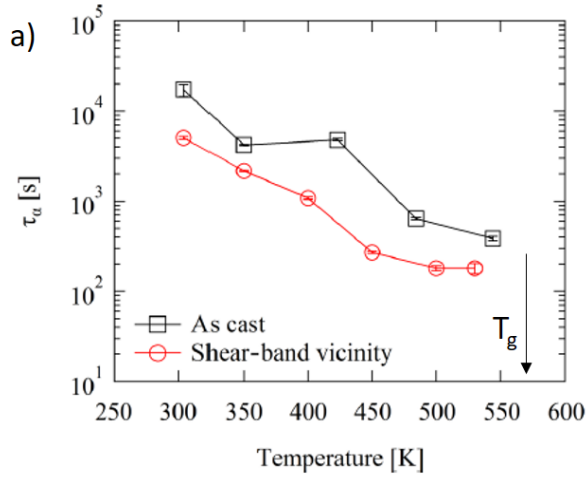

Figure 6. Temperature dependence of characteristic time τ obtained from site-specific isothermal XPCS measurements at 10 μm from an individual shear band and in the bulk in a $Zr_{65}Cu_{25}Al_{10}$ MG (as-cast from suction casting, stepwise heating from the ambient temperature). The significant decrease of τ in the shear band vicinity demonstrate an effect in an area much larger than the shear band itself. Figure taken from [49].

Interestingly, the acceleration of the dynamics remains constant during heating from room temperature to 0.95×Tg (Fig. 6a). This indicates that the structural and dynamical changes due to the shear band do not relax back to the as-cast (or annealed) state during the experimental time even close to T_g, and that the faster dynamics could persist over a very long time at ambient temperature, enabling true control of the glassy dynamics.

The fast dynamics near a single shear band described above occur also in the case of a severely deformed bulk MG. Results from XPCS performed on $Pd_{40}Ni_{40}P_{20}$ glasses annealed and later deformed through high pressure torsion (Fig. 6b) show a trend nearly identical to the single shear band case before structural relaxation close to T_g ("Crossover" zone in the Fig. 6b) [50].

Different deformation protocols not triggering the formation of shear bands can also induce rejuvenation or affect the dynamics. Hydrostatic compression was shown to allow large volumetric changes without the development of shear bands, and leads to a significant rejuvenation visible from calorimetry measurement [51]. How this translates to the atomic scale dynamics compared to sheared samples remains an open question. On the other hand, while the deformation of a metallic glass below the yield stress of the material do not produce permanent modifications by definition, XPCS performed in-situ during the elastic bending of $Zr_{44}Ti_{11}Cu_{10}Ni_{10}Be_{25}$ glassy ribbons shows a deformation influence on the dynamics that depends on the thermal history of the sample [30]. On the as-cast samples quenched from the

melt by rapid cooling, the deformation increases the intermittent nature of the aging. On the other hand, when the sample has been brought to a lower energy state by pre-annealing just below T_g, the deformation leads to faster but steady dynamics.

All these works show how the relaxation dynamics of MGs can be tuned by different experimental protocols, which can involve either the glass formation or their treatment. While physical vapor deposition can effectively bring the glass in lower energy microscopic configuration associated to larger barriers and longer times for structural rearrangements, applied stresses and deformations rejuvenate the system, bringing the glass in a microscopic state characterized by fast structural relaxation times but at the same time more homogeneous dynamics with respect to physical aging. Further studies will be necessary to better clarify the controlling physical mechanisms controlling the collective particle motion.

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