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Report:

Bulk metallic glasses (BMGs) are currently among the most actively studied glasses and metallic materials. However, like all glasses, BMGs exhibit physical aging and thus any observable spontaneously evolves with time and displays memory effects related to the previous history of the material. For their successful use in commercial applications, it is therefore necessary to gain detailed knowledge of the physical mechanism responsible for aging in BMGs, which is essential at a technological level, e.g. for controlling material properties. Despite extensive studies of the evolution of macroscopic properties during aging, such as enthalpy, viscosity, elastic modului, fracture toughness etc., still very little is known about the atomic-scale mechanisms responsible. Recently, XPCS has been successfully applied to follow for the first time the temporal evolution of the structural relaxation time, τ, in BMGs at the atomic length scale opening the field to a vast range of investigations [1].

In Experiment number HC-1422, we conducted XPCS experiments to measure the atomic relaxation dynamics in $Zr_{44}Ti_{11}Ni_{10}Cu_{10}Be_{25}$ and $Pd_{43}Cu_{27}Ni_{10}P_{20}$ bulk metallic glass (BMG) compositions. The specific focus of these studies was to follow the evolution of the structural relxation process from the glassy state into the equilibrium undercooled liquid by means of isothermal annealing. We applied thermal protocols identical to those of previous relaxation studies where the equilibration of macroscopic properties (e.g. enthalpy, volume and viscosity) was measured. By comparing the results measured on the atomic-scale with the macroscopic data, we aim to establish a better understanding of the microscopic mechansim involved in aging in BMGs, as well as to test the validity of the Maxwell relation, $\eta=G_\infty\tau$, where G_∞ is the infinite-frequency shear modulus

Measurements were carried out on as-spun samples of amorphous BMG ribbons, which were held in a resistively heated furnace, evacuated to a pressure < 10^{-2} mbar. After heating into the supercooled liquid, the samples were cooled down with a rate of 1 K/min and time series of up to 5000 images were taken with 5 s exposure per frame at the specified holding temeprature. Figure 1 shows representative two-time correlation functions $G(t_1, t_2)$ during isothermal aging of $Zr_{44}Ti_{11}Ni_{10}Cu_{10}Be_{25}$ (left) and $Pd_{43}Cu_{27}Ni_{10}P_{20}$ (right). The width of the reddish intensity profile along the main diagonal gives information on the structural relaxation time and its temporal evolution during the isothermal measurement. In both samples we observe a steady broadening of the intensity contour with time which indicates an increase of the relaxation time and thus a slowing down of the dynamics.

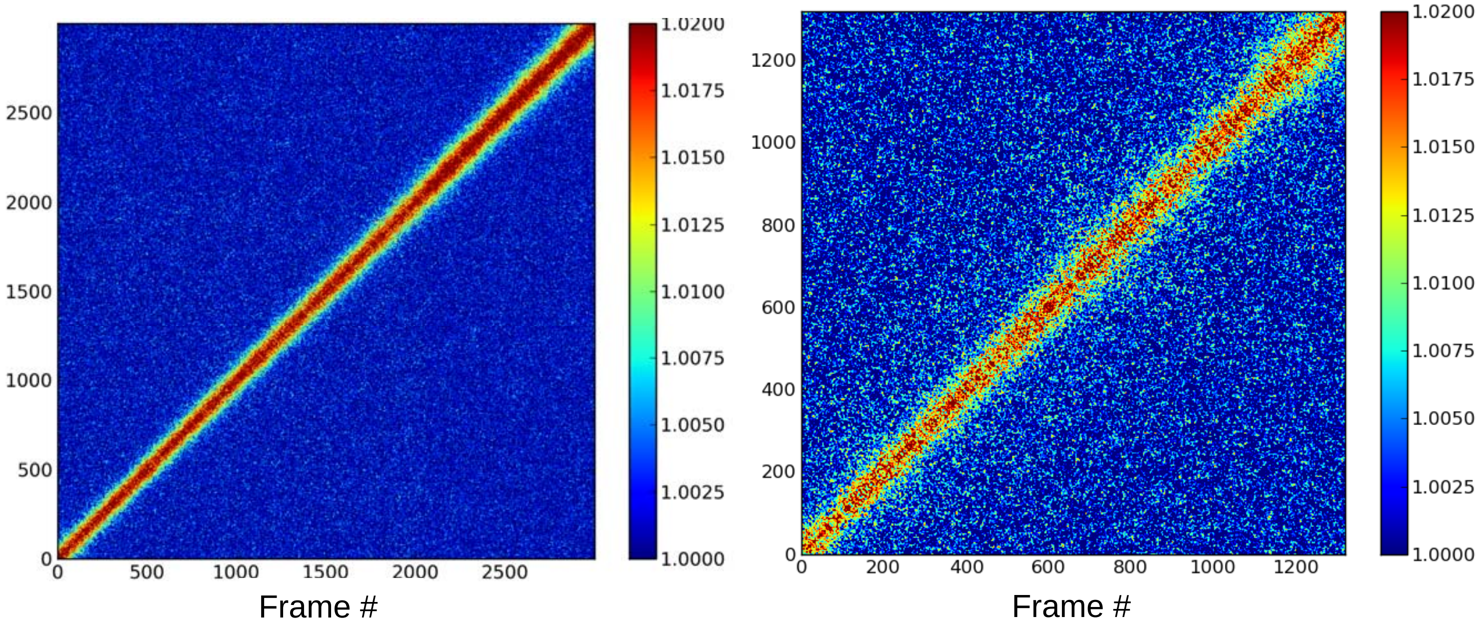

Figure 1 (left) $G(t_1, t_2)$ of $Zr_{44}Ti_{11}Ni_{10}Cu_{10}Be_{25}$ during aging at T = 312 °C and **(right)** $Pd_{43}Cu_{27}Ni_{10}P_{20}$ during aging at T = 280 °C. The axes have the units of frames and the exposure time for each sample was 5 s.

In total, 52 individual measurements were performed for $Pd_{43}Cu_{27}Ni_{10}P_{20}$ and 15 for of $Zr_{44}Ti_{11}Ni_{10}Cu_{10}Be_{25}$. The equilibration of the structural relaxation time as a funciton of the waiting time – extracted from the $G(t_1, t_2)$ – was monitored for selected temperatures, for which macroscopic equilibration data was available. Measurement were also carried out in the supercooled liquid region, above T_g; however, reliable data were scarce in this region due to insufficient temporal resolution at shorter times. Additional measurements were carried out at room temperature for both ribbon and bulk samples. So far, the initial focus has been devoted to analyzing each of the collected data sets. Our ultimate goal is to establish a better understanding of the atomic mechanisms involved in structural relaxation and aging in BMGs.

[1] B. Ruta, Y. Chushkin, G. Monaco, L. Cipelletti, E. Pineda, P. Bruna, V. Giordano, and M. Gonzalez-Silveira, Phys. Rev. Lett. **109**, 165701 (2012).