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Atomic Motion in Metallic Glass Studied by Coherent X-Rays

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1. Introduction

It is widely accepted that understanding the temperature-driven motion of single atoms in solids is more complicated than comprehending their structural properties. Understanding the atomic transport in a glassy state seems to be a still more difficult task. There are numerous simulations testing the dynamical properties of glassy systems but experimental works are rather scarce. Tracer methods have been applied to study diffusion in various metallic glasses [1], being, however, limited to purely macroscopical examination of considered systems. Probing the dynamics with spatial and temporal resolution sounds very promising but turns out to be an extremely challenging task due to the behavior of the glass formers, particularly the dramatic slowing down of the dynamics in systems undergoing a glass transition. A lot of research efforts have focused on the study of colloidal systems with multispeckle dynamic light scattering [2] and with X-ray photon correlation spectroscopy (XPCS) [3]. Especially the last method seems to be very promising in the study of dynamics with atomic resolution as it has been just demonstrated in the intermetallic alloy $Cu_{90}Au_{10}$ [4].

Although studies of colloidal systems are experimentally feasible compared to tough measurements on metallic glasses especially due to the orders of magnitude stronger scattering intensities, metallic glasses posses, however, certain advantages compared to molecular glass formers: metallic glasses can be in good approximation regarded as multicomponent hard-sphere-like systems, evidently easier to understand than delicate and preparation-history dependent molecular structures. We hope that a general description of hard-sphere dynamics should be subsequently applicable to a wide variety of amorphous materials.

2. Time-Resolved XPCS Measurements

The experiment was carried out at beamline ID10A at the European Synchrotron Radiation Facility in Grenoble, France. The wavelength of applied radiation was 1.55 $Å^{-1}$. The sample was mounted in a vacuum furnace in transmission geometry. Scattered radiation was detected using a direct illumination CCD camera (1024×1024 pixels). Time series were interpreted in terms of the intensity auto-correlation function $g^{(2)}(q, \Delta t)$ with q being the scattering vector and Δt the time-lag between intensities in one pixel.

The sample under investigation was a Zr-based amorphous Inoue alloy [5] $Zr_{65}Al_{7.5}Ni_{10}Cu_{17.5}$ with a calorimetric glass transition T_g of 624K (measured with heating rate of 2K/min) and with extrapolated quasi-stationary T_g of 605K. The sample was pre-annealed at 600K for 25h and additionally 15h at the beamline before the accumulation of the data at different scattering angles at 600K took place. During in situ annealing the

data were also accumulated and evaluated via a two-time correlation formalism ensuring achievement of the quasi-stationary condition before the XPCS measurement was started. Finally the time series were measured for three scattering angles $2\theta = 32°$ ($q = 2.23$ Å^{-1}), 37° (2.57 Å^{-1}) and 42° (2.90 Å^{-1}), where $2\theta = 37°$ is the position of the first diffuse peak. Fitting the data was possible only via a compressed exponential function $g^{(2)}(q, \Delta t) - 1 = \exp[-(\Delta t/\tau)^{\beta}]$, where $\tau(q)$ denotes a correlation decay time and β is the universal compressing exponent $\beta = 1.8(1)$, compare solid lines in Fig. 1.

3. Conclusion

One should notice that the fundamental characteristics of the XPCS method resemble that of quasi-elastic neutron scattering and are different from resonant methods like Mössbauer spectroscopy. The decay time $\tau(q)$ which is the life-time of fluctuations corresponding to the wave-vector q, thus, gauges the dynamics in the sample. XPCS is a method sensitive to the pair-correlation function which makes this method especially sensitive to collective excitations generally supposed to be operative in glassy systems [1].

The measured q-dependence of the decay time $\tau(q)$ negates the role of vacancies in glassy dynamics at the measurement temperature. A thorough consideration of the two-time correlations during equilibration of the system suggests the role of heterogeneous dynamics found in most molecular glass formers [3].

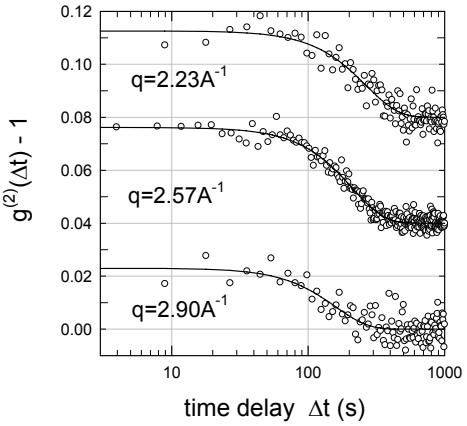

Fig. 1: Intensity correlation function at various q's and $T = 600$ K. Data points are shifted vertically relative to each other by 0.04 for clarity and well described by a compressed exponential function.

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