ESRF	**Experiment title:** Partial Vibrational Density of Phonon States in NANOPERM $Fe_{90}Zr_7B_3$ nanocrystalline alloy seen by Nuclear Inelastic Scattering of Synchrotron Radiation	**Experiment number**: MA-83
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Report:

In this experiment we have studied the density of phonon states (DOS) in NANOPERM-type nanocrystalline (NC) alloy via nuclear inelastic scattering (NIS) spectroscopy aiming to separate the DOS of atoms located at the grains' interior from those occupying the grain boundaries (GB). The experiment was motivated from recent computer simulations based on the Density Functional Theory (DFT) which were able to separate the DOS of the nanograins from those of the GB in nanocrystalline Ni, revealing unusual vibrational properties of the atoms located at the GB [1]. An amorphous ribbon with stoichiometric composition of $^{57}Fe_{90}Zr_7B_3$ enriched to 65% to the resonant isotope ^{57}Fe was prepared by rapid quenching in a vacuum by the melt-spinning technique. The controlled annealing of the amorphous precursor results in formation of nano-crystals of α-Fe with *bcc* structure having well-developed GB areas. Fig. 1 summarises the extracted DOS (a) and reduced DOS (b) of the NC samples prepared at the indicated conditions, together with those of the amorphous precursor and 80 μm thick α-Fe foil. Precise analysis of the experimental DOS confirmed a quadratic, Debye behaviour of the low-energy part down to 1 meV for all measured spectra. Singular phonon excitations at the high-energy part of the DOS, predicted by the DFT calculations, have not been observed. As reported earlier damping of the longitudinal peak at 35 meV as a function of the grain-sizes has been confirmed [2].

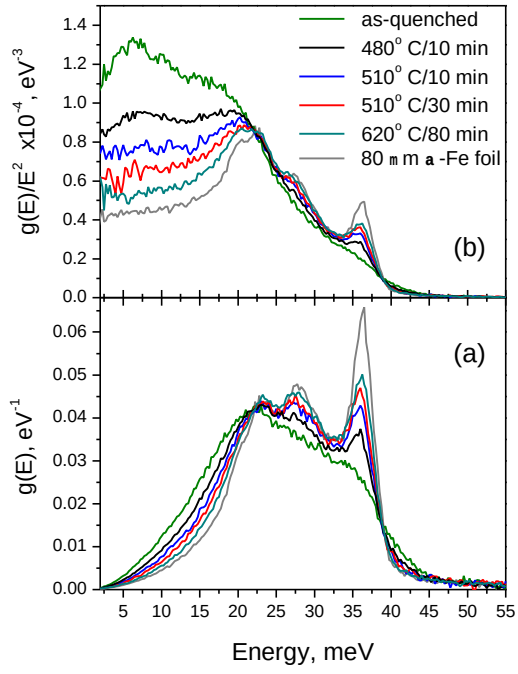

Fig.1 DOS (a) and reduced DOS (b) of nanocrystalline $^{57}Fe_{90}Zr_7B_3$ alloy prepared by annealing at the indicated conditions, together with DOS of the amorphous precursor and a-Fe foil.

Fig. 2 shows a typical conversion electron Mössbauer spectrum (CEMS) of a NC $^{57}Fe_{90}Zr_7B_3$ alloy prepared by annealing at 510°C for 10 min. One can clearly separate a narrow sextet indicating a perfect *bcc* crystalline phase (CR), a broad sextet associated with the interface atoms (IN)

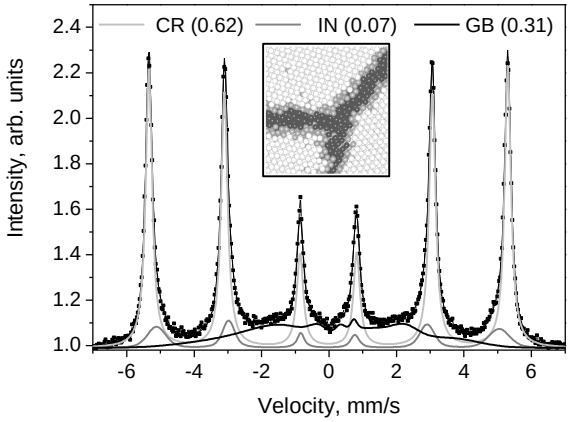

Fig. 2. CEMS spectrum of NC sample prepared at 510 ℃ for 10 min. The inset shows a model of the system with the CR, IN and GB phases associated with the Mössbauer sub-spectra.

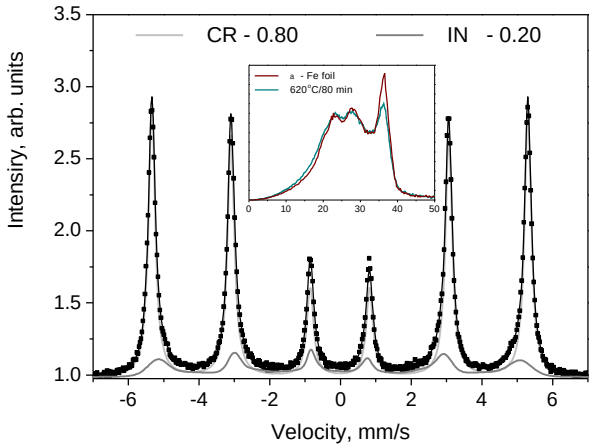

Fig. 3. CEMS spectrum of the sample prepared at 620 ℃/80 min. The inset shows the DOS of this sample compared with the DOS of α-Fe foil.

and broad sub-spectrum indicating a rather disordered phase originating from the grain boundaries. The inset shows a model of the studied NC system, where one can distinguish the corresponding three phases - CR (white), IN (grey) and GB (black) atoms, associated with the Mössbauer sub-spectra. The first attempt to fit the experimental DOS with the function:

$g_{fit}(E) = Ag_{a\text{-}Fe}(E)*L(G) + (1\text{-}A)g_{Am}(E)$, with $g_{\alpha\text{-}Fe}(E)*L(G)$ being the DOS of α-Fe foil convoluted with a Lorenzian with FWHM G and $g_{Am}(E)$ the DOS of the amorphous precursor, resulted in a relatively large misfit. The main reason for the discrepancy is the missing DOS of the interface component in the used model. However a closer inspection of the CEMS of the sample treated at 620°C/80 min, Fig. 3, shows well-crystallised state with negligible presence of the disordered component. The inset shows the DOS of this sample compared with the DOS of α-Fe foil. The main difference is the damping of the peak at 35 meV and the excess of phonon states at low energies, which is to be expected for the atoms located at the interfaces [1]. Therefore we used the DOS of that sample as a model for the CR+IN phases in the NC samples and their DOS were fitted with the function:

$g_{fit}(E) = Ag_{620C/80min}(E)*L(G) + (1\text{-}A)g_{Am}(E)$, A and G being fit parameters. Results for the measured samples are plotted in Fig. 4, together with the partial DOS of the CR+IN and the GB components. The derived partial areas are in very good agreement with those obtained from the CEMS measurements. Complimentary XRD studies for precise determination of the grain sizes are in progress.

In conclusion:

- *For the first time the DOS of the GB in nanocrystalline systems have been experimentally identified. It was unambiguously shown that the DOS of the GB can be fully described by the DOS of disordered, amorphous phase.*
- *For all studied samples the low-energy tail of the DOS is found to exhibit the quadratic Debye behaviour.*
- *Despite of the high statistical quality of the collected NIS spectra peculiar phonon excitations in the energy range between 45 and 50 meV associated with the GB have not been detected.*

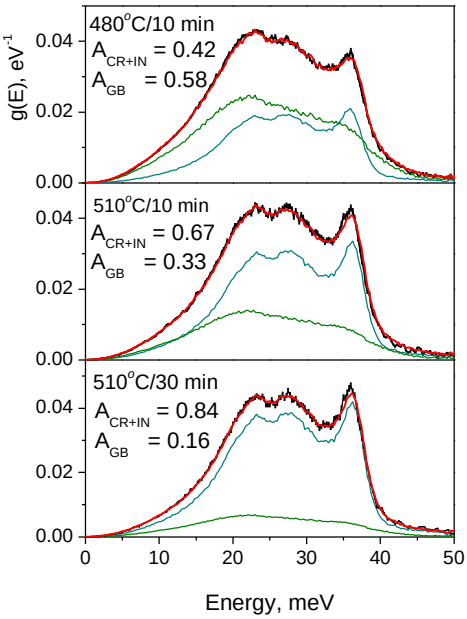

Fig. 4. The experimetal DOS of the samles prepared at the indicated conditions, togeter with the fit to the function $g_{fit}(E)$.

[1] P.M.Derlet et al., PRL **87** 205501 (2001), **92** 035505 (2004).
[2] L. Pasquini et al., PRB **66** 073410 (2002).