



	Experiment title: Disentangling atomic and mesoscopic dynamics of disordered solid: can one boson peak hide another?	Experiment number: HC-1659
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Report:

The aim of this study was to disentangle the atomic dynamics of disordered solids related to atomic and to mesoscopic scales. Most theoretical models attribute the excess of heat capacity and the related excess of vibrational states of glasses (i.e., the “boson peak”) to glass-specific phenomena caused by disorder and occurring at mesoscopic length scale. Our recent experiments, however, suggest that they are related to short-range atomic structure, that they are the counterparts of the well-known phenomena occurring in crystals (i.e., acoustic van-Hove singularity), and as such are more reflecting residual order presented in glasses [1, 2].

The exact interpretation, however, still remains a matter of belief, because typical glasses are built out of large structural units. In this case, the residual order is characterized by the same mesoscopic (~nm) length scale and, therefore, both effects appear at nearly the same energy. In order to disentangle them, here we studied an amorphous iron – disordered solid with very small (atomic size) structural unit. For this system, the mesoscopic and atomic dynamics should be separated in energy by almost an order to magnitude and, therefore, should be easily distinguishable from each other.

We applied for beamtime at ID28 in order to study the dispersion relations and at ID18 for measurements of the density of vibrational states. Our application was granted only by beamtime at ID28, therefore, here we report on the measurements of the dispersion relations.

We studied two samples (A and B) of amorphous iron prepared by micro-chemical reaction [3], one sample (C) of amorphous iron obtained by sonication [4], and we also measured polycrystalline iron powder sample (D) for comparison. The samples A and B were preliminary characterized by X-ray diffraction, which confirmed their amorphous state (Fig.1). The sample C was prepared just before the experiment. Its amorphous state was verified in-situ, during the measurements of dispersion relations (see below).

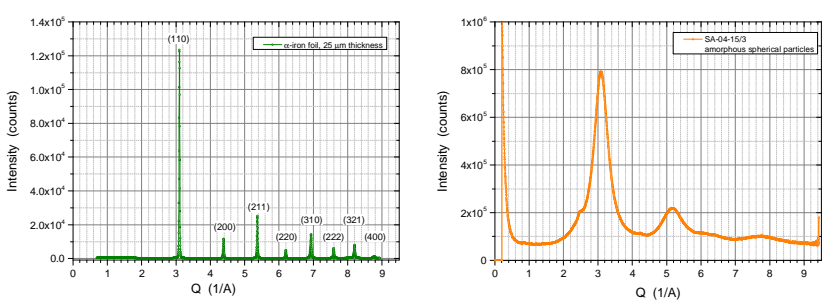
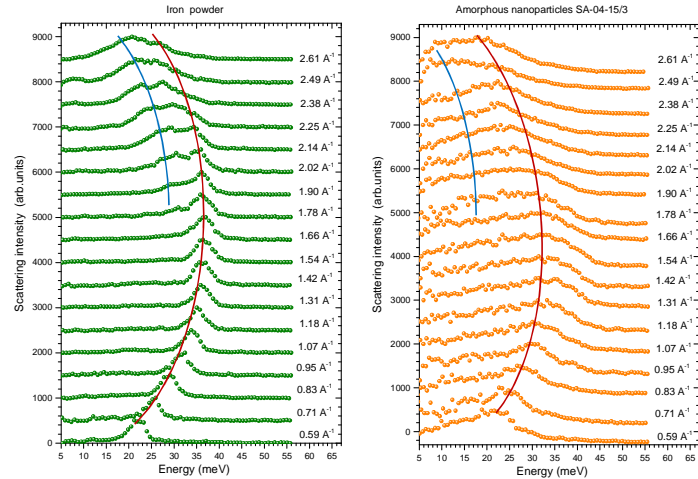


Fig.1. X-ray diffraction data for α -iron powder (left) and for amorphous iron (sample A). The fully amorphous nature of the sample A is revealed by the absence of sharp diffraction peaks. Similar pattern was also observed for the sample B.

Figure 2 shows the dispersion relations measured for iron powder (sample D, left panel) and for amorphous iron (sample A, right panel). The iron powder sample (left panel) reveals known dispersion relations for α -iron. Within the first Brillouin zone ($Q < 1.5 \text{ \AA}^{-1}$), according to the selection rules, only the longitudinal branch is seen. In the second Brillouin zone ($Q > 1.7 \text{ \AA}^{-1}$), the transverse acoustic excitations also appear.

The dispersion relations for amorphous iron (Fig.2, right panel) show very similar behavior, although here the excitations are strongly broadened and shifted to lower energy. Possibly, even the transverse excitation could be traced in the second pseudo-Brillouin zone. This, however, requires more detailed analysis.

Fig. 2. The measured dispersion relations for α -iron powder (left panel) and for amorphous iron (sample A, right panel). In both cases, the dispersive behavior of acoustic excitations is clearly seen. The red curve emphasizes the dispersion of the longitudinal branch, whereas the blue curve indicates the dispersion of transverse excitations, which is clearly seen for α -iron (left column) and can possibly be also traced for amorphous iron (right panel). Similar pattern was observed also for the amorphous sample B.



The measured dispersion relations for amorphous iron suggest that the corresponding density of states (DOS) should have a strong peak at the energy of about 10-15 meV, which corresponds to piling-up of the transverse acoustic excitations near the boundary of the pseudo-Brillouin zone ($Q = 1.5 \text{ \AA}^{-1}$). Furthermore, one may also expect a smaller peak or a shoulder at the energy of about 25-30 meV, which would correspond to the similar piling-up of the longitudinal excitations. These suggestions, as well as the presence of possible peaks at lower energy, which would then correspond to the mentioned above effects in mesoscopic dynamics, should be verified in measurements of the DOS.

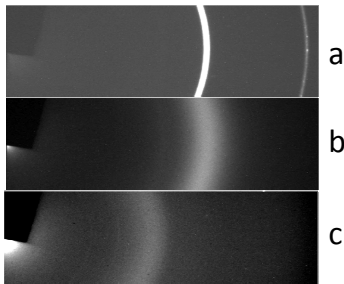


Fig.3. The diffraction patterns measured during the in-situ control of the samples in dispersion relation measurements for (a) iron powder, sample D, (b) for the amorphous sample A, and (c) for the amorphous sample C.

In addition, we note that the in-situ control of the samples in measurements of the dispersion relations revealed an unexpected diffraction pattern for the amorphous sample C. In contrast to samples A and B, where the broad halo appears at the Q-vector close to the (110) reflection of α -iron, the scattering pattern for the amorphous sample C reveals a ring at approximately 40% smaller Q-vector, which corresponds to $\sim 2.8 \text{ \AA}$ distance between the nearest neighbors (Fig.3). For crystalline iron, such a peak would correspond to a *primitive cubic cell structure*, which was never observed for iron crystals. Thus, the sample C could be a new polymorph of amorphous iron with an exotic short range order. In measurements of the dispersion relations, the inelastic fraction of scattering for this sample was completely hidden by the tails of the dominating elastic central peak. However, we hope to measure the DOS for this sample using a high energy resolution of IXS with nuclear energy analysis at ID18, and we also will study X-ray diffraction for this sample.

References:

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