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Received by 0001

OCT 0 4 1991

TITLE: Neutron Brillouin Scattering In A Metallic Glass

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SUBMITTED TO: Journal Of Non-Crystalline Solids
(Proceedings of NCM-5, Sendai Japan, September 1991)

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The dispersion of collective modes in a metallic glass ($\text{Mg}_{70}\text{Zn}_{30}$) measured earlier at the thermal neutron time-of-flight spectrometer IN4 of the HFR of the ILL could be extended towards lower momentum transfers down to the first pseudo-Brillouin zone for the first time. This extension to momentum transfer not accessible up to now was possible using the highly resolving time-of-flight spectrometer HET of the new spallation source ISIS. In the region of overlap the two parts of the dispersion determined with different samples of the same metallic glass on different instruments agree very well. Also the earlier discrepancies with the dispersion determined for this metallic glass from a computer simulation could be nearly completely eliminated due to a more recent and more complete investigation of this glass.

1. INTRODUCTION

For an infinite periodic crystal a reciprocal lattice with infinitely many equivalent Brillouin zones can be defined. Thus, in principle, one can measure the dispersion of phonons in every of those Brillouin zones, even though in reality the structure factor of the phonons (scattering intensity) is normally higher at larger momentum transfers $\hbar Q$ and larger Q -values are therefore preferred in most of these experiments.

For disordered matter no reciprocal lattice exists, but as long as these systems have a pronounced short range or even medium range order like they exist in dense fluids and simple topologically disordered solids, low order pseudo Brillouin zones can be defined near the origin of reciprocal space, in which then the first pronounced maximum at Q_p of the static structure factor $S(Q)$ plays the role of the first (smeared out) reciprocal lattice point in a reciprocal lattice. In fact, this rather vague definition of a pseudo Brillouin zone can be justified *a posteriori* also by the shape of the measured dispersion of collective atomic motions, as is demonstrated by the results presented here. According to this analogy the first Brillouin zone should extend up to $Q_p/2$ in the reciprocal space of these samples, which are assumed to be isotropic. In analogy to light scattering from acoustic phonons in crystals, where even the largest Q values do not exceed $\sim 0.04 \text{ nm}^{-1}$, neutron scattering experiments with momentum transfers below $\hbar Q_p/2$ have been called neutron Brillouin scattering (NBS) experiments [1].

While for single crystals such experiments are of interest only in very few cases, for disordered material NBS experiments are of primary importance for the investigation of their collective atomic dynamics. If the lifetime of the collective atomic vibration is long enough (e.g. some ps) these excitations can usually be seen as peaks in the inelastic part of the dynamic structure factor $S(Q, \omega)$ similar to the peaks caused by phonons in the inelastic neutron scattering spectra, but normally they are much less intense and broader than the phonon peaks. The width of these peaks increases rapidly with increasing

momentum transfers. Thus at small Q -values these peaks are not only easier to observe but one would also have a chance to determine the intrinsic width of these peaks, which is related to the lifetime of the excitation and with this to the physical processes which lead to the decay of the collective atomic vibration.

A second argument in favour of measuring the collective atomic dynamics of a topologically disordered solid at small momentum transfers, is given by the fact that at higher momentum transfers, usually at $Q > Q_p$, the structure of the inelastic part of $S(Q, \omega)$ is mainly determined by the structure of the vibrational density of states and with this by the single particle motion in the amorphous solid.

A third advantage of NBS experiments is given by the fact that neutrons couple directly to longitudinal vibrations while they couple to transverse vibrations via Umklapp processes outside the first Brillouin zone. Thus NBS experiments give the possibility to separate longitudinal excitations from transverse excitations.

In spite of these strong motivations to use NBS experiments to investigate the dynamics of amorphous solids, to our knowledge no experiment of this kind has been done up to now. The reason for this is that the experimental requirements of NBS experiments are very demanding and could hardly be realized for systems for which the slope of the dispersion curve of the excitations under investigation corresponds to sound velocities of 2 to $7 \cdot 10^3$ m/s. Only with the advent of highly resolving (1-3% in energy) time-of-flight (TOF) spectrometers at the new spallation sources, NBS experiments on solids have become possible. Here we report on the first of these experiments done on the metallic glass $Mg_{70}Zn_{30}$.

One of several contradicting requirements is that the velocity v_0 of the incident neutrons has to exceed the velocity of the excitation under investigation by a factor of 1.3 to 1.5, i.e. the dispersion curve has to lie well within the dynamical range of the experiment (energy- and momentum transfers covered). With increasing incident

however, the energy resolution of the spectrometer strongly deteriorates. Thus the peaks in the inelastic part of $S(Q, \omega)$ can no longer be resolved. At the same time, the Q -value measured at a fixed scattering angle θ increases with increasing E_0 .

$$Q = \frac{\sqrt{2m}}{\hbar} (E_0 + E - 2\sqrt{E_0 E} \cos \theta)^{1/2}. \quad (1)$$

Thus with high E_0 , one has to work at smallest scattering angles in order to reach Q values below $Q_p/2$.

2. EXPERIMENT

The melt spun material was kept in a flat Al frame with a sample volume of $6 \times 6 \times 2 \text{ cm}^3$. As the amount of multiple scattered neutrons is very high compared to the low intensity of single scattered neutrons in NBS experiments this frame was intersected horizontally and vertically by 0.5 mm thick cadmium plates (very strong neutron absorber), 2 cm wide, in order to absorb the neutrons scattered several times in the sample. The compressed sample material was put in the $1 \times 1 \times 2 \text{ cm}^3$ openings of this grid and the frame was then closed by 0.3 mm thick Al foils at both sides of the frame. An identical container was prepared for the empty can measurement and a similar sample holder without Cd intersections was assembled for the 2 mm Vanadium plate used for the calibration runs at the beginning and at the end of the experiment. For the Cd runs the empty container was covered with 1 mm Cd on each side of the frame. During the experiment the sample holders were covered in Boron frames, reducing the frame part accessible for neutrons to the beam size of $4.8 \times 4.8 \text{ cm}^2$.

The experiment was done at room temperature at the high energy transfer TOF spectrometer HET at the spallation source ISIS of the Rutherford Appleton Laboratory. Because the absorption maximum of Cd is at 180 meV, an incident energy of 180 meV was chosen in order to maximize the effectiveness of the Cd intersection of the sample. The detectors are fixed with axial symmetry around the through-going primary neutron

beam on two sections of spheres with 4 m and 2.5 m radius, the sample being in the common centre of these spheres. The 50 detectors at 4 m distance were grouped to 10 concentric rings at scattering angles between 3.3 and 7 degrees. Finally, in order to improve the statistical accuracy, 3 rings of this kind were added together. The detectors of the 2.5 m flight path section were grouped into 32 rings covering scattering angles between 9.3 and 28.7 degrees. For elastic scattering ($E = E_0$ in equ. (1)), this corresponds to momentum transfers $Q(E_0)$ between 7 and 46 nm^{-1} as can be seen in Fig. 1, where the static structure factor of our sample is shown, calculated by integrating $S(\theta, \omega)$, the dynamic structure factor measured at fixed scattering angle θ . In transforming the measured intensities to absolute values of $S(\theta, \omega)$ and $S(Q)$ with help of the two V-calibration runs, the lack of intercalation of the V-sample (no shading of the sample) had to be compensated by an additional general normalization factor of 1.7. The resolution of the spectrometer at $\omega = 0$, as obtained from the sum of all elastic lines in the V-spectra measured with the detectors in 4 m distance, was 5 meV. At 2.5 m flight path the resolution is worse.

3. DATA TREATMENT AND RESULTS

The raw data were treated in two different ways in order to determine the energy corresponding to the peaks in the inelastic part of $S(\theta, \omega)$.

After a simple subtraction of the measured background, caused by scattering from the sample holder and windows in the spectrometer, a two channel maximum entropy (TCME) method was applied to separate the more pronounced structure in the inelastic neutron scattering spectra of the sample from the broad distribution of multiple scattered neutrons. Separation between the structured and the broad part of the spectrum is possible due to constraints in the two channels. One channel is restricted to accept a broad structure only, the other one is allowed to accommodate a sharply structured spectrum, if

this is contained in the data [2]. Fig. 2 shows the result (the structured part) of this TCME reconstruction of the spectrum measured with the detector ring at $\theta = 9.3$ degrees of the 2.5 m flight path. The TCME method is applicable as long as the measured spectrum can be clearly separated in a part with pronounced structure and one which is treated as a broad background. As is shown in Fig. 2, the position of the side peaks can easily be determined in the reconstructed spectrum.

In the second data treatment the symmetrized dynamic structure factor $S(\theta, \omega)$ was determined on absolute scale, calculating all necessary corrections except those for the finite resolution of the spectrometer and for multiple scattered neutrons [3]. The $S(Q)$ calculated by integrating $S(\theta, \omega)$ showed that about 70% of the intensity measured at lowest scattering angles in the detectors at 4 m distance was due to multiple scattered neutrons. This spectrum therefore was fitted by a spline function (in order not to deteriorate the anyhow low statistical accuracy of the data) and 70% of this spectrum was subtracted from all subsequent spectra. After this crude correction for multiple scattered neutrons, which introduces an additional error for the absolute intensity of the spectra, the resulting $S(\theta, \omega)$ is shown in Fig. 3 again for the spectrum measured at $\theta = 9.3$ degrees and 2.5 in flight path (compare Fig. 2).

4. DISCUSSION

Plotting the energies E_m corresponding to the maxima in the neutron energy gain and loss part of $S(\theta, \omega)$ (left and right to the foot of the elastic peak in Fig. 3) in dependence of the corresponding momentum transfer $\hbar Q(E_m)$, one can derive a dispersion curve of the collective excitations in the metallic glass. In Fig. 4 the present results (●, = with error bars) are compared with the results of an earlier experiment [4] done at the thermal neutron TOF spectrometer IN4 of the ILL, in which only the part of the dispersion near Q_p and at Q_p [5] outside the first Brillouin zone could be measured

due to instrumental limitations at that time. Despite the fact that the dispersion between 17 and 23 nm⁻¹ (IN4) and the dispersion between 12 and 21 nm⁻¹ (HET) were measured with different samples of the same metallic glass under completely different experimental conditions, the results agree very well in the region of overlap of these two independent experiments. For reasons of consistency, in this comparison the energies E_m obtained from $S(\theta, \omega)$ have been used in the case of the earlier IN4 experiment, which are a few percent higher than the E_m values obtained from $S(Q = \text{const}, \omega)$, published in ref. [4]. The dispersion derived from the peaks in the inelastic spectra after the TCME reconstruction (see Fig. 2) agrees within the error bars with the dispersion shown in Fig. 4.

The crosses in Fig. 4 represent the energies corresponding to the peaks in the calculated total dynamic structure factor $S(Q, \omega)$ for the metallic glass Mg₇₀Zn₃₀ [6] which was obtained as a weighted sum from the calculated partial $S_{ij}(Q, \omega)$

$$\sigma S(Q, \omega) = 4\pi \sum_i \sum_j b_i b_j c_i c_j S_{ij}(Q, \omega) \quad (2)$$

where b_i , c_i are the coherent neutron scattering length and the concentration of the element i in the binary alloy. In the earlier comparison the measured dispersion relation disagreed with the calculated one by nearly a factor of 2 and led to the misinterpretation of the peaks as being due to transverse excitations. In contrast to this, the results of the present and the earlier experiment agree rather well with these new calculations. In addition, in the present experiment some excitations were measured in the first pseudo Brillouin zone. Thus the interpretations of these excitations being due to longitudinal modes is justified.

Acknowledgement

We would like to thank Prof. H.J. Güntherodt for making available the MgZn sample, H. Breitenstein for the melt spinning of the sample, and the ISIS facility for providing us with beamtime. It is a pleasure to acknowledge the technical assistance obtained at the HET spectrometer by Z. Bowden and T. Jones.

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Figure Captions

- Fig. 1 : Static structure factor $S(Q)$ of the metallic glass $Mg_{70}Zn_{30}$ as obtained from the integration of $S(\theta, \omega)$ determined in this experiment. Q_p is at 26 nm^{-1} . Of the 10 detector rings at 4 m flight path only the two groups of 3 rings (●) were finally used. From the 31 detector rings of the 2.5 m flight path (○) only the 6 at lowest scattering angles have contributed to Fig. 4.
- Fig. 2 : Maximum entropy reconstruction of the structured part of the spectrum measured at a scattering angle θ of 9.3 degrees and with a flight path of 2.5 m for the metallic glass $Mg_{70}Zn_{30}$. The broad part of the spectrum, assigned to multiple scattered neutrons, is not shown.
- Fig. 3 : $S(\theta, \omega)$ of the metallic glass $Mg_{70}Zn_{30}$ measured at a scattering angle of 9.3 degrees and with a flight path of 2.5 m. The excitations measured in neutron energy gain and neutron energy loss are clearly seen left and right of the foot of the elastic peak (only its foot is shown in order to augment the inelastic part of $S(\theta, \omega)$).
- Fig.4 or 5 : Energies $\hbar\omega$ corresponding to the maxima in the inelastic part of $S(\theta, \omega)$ measured at const. scattering angle θ . ● : Neutron energy loss side, ■ : neutron energy gain side of the spectra. ○, • : corresponding results from an earlier investigation of the dispersion of collective excitations of the same metallic glass [ref. 4]. X : energies corresponding to the maxima in the calculated total dynamic structure factor [ref. 6]. The broken line is thought as a guide for the eyes through these crosses.

Fig. 1

Fig 3

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