Phonon dispersion of metallic glass CuZr₂

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Abstract. Collective dynamics of metallic glass CuZr₂ has been studied in the first pseudo Brillouin zone using high-resolution inelastic X-ray scattering. Acoustic-like longitudinal propagating excitations were observed and the dispersion relation was determined. In addition of longitudinal mode, transverse mode with half excitation energy contributes to medium energy-transfer region.

1. Introduction

It is known that CuZr and NiZr alloys can form metallic glass by rapid quenching from high-temperature liquid. Fukunaga et al.[1] revealed that Cu and Zr atomic configuration in this metallic glass is likely to that of dense random packing model, where the nearest neighbor coordination is almost 12. The study of static structure and glass forming mechanism focuses into regular icosahedral cluster which has 12 equivalent vertices and 5-fold symmetry, the latter of which prevents nucleation in the liquid. On the other hand, the structure of NiZr₂ alloy is somehow different from the random close packing model where prism-type polyhedrons frequently appears in Voronoi polyhedron analysis on the structural model generated by reverse Monte Carlo method.

Among the Zr based metallic glass, high frequency dynamics of NiZr₂ alloy are extensively investigated by neutron [2] and X-ray inelastic scattering measurements [3]. The analysis revealed that longitudinal acoustic phonon exists and the phonon dispersion relation shows pseudo Brillouin zone where zone centers lie approximately at the maxima in the static structure factor. Because of much higher stability of glass state in CuZr₂ than in NiZr₂, investigation of high frequency excitations for
CuZr$_2$ metallic glass is indispensable to understand collective dynamics of metallic glass. We present the results of inelastic X-ray scattering (IXS) measurements for metallic glass CuZr$_2$ and the phonon dispersion relation.

2. Experimental procedure
Development of inelastic X-ray scattering instruments allows us to explore the high-energy-transfer region at a relatively low momentum-transfer where phonon-like propagating modes exist. The experiments were carried out at the beamline BL35XU [4] of SPring-8, Hyogo, Japan using a horizontal IXS spectrometer. The energy resolution of the spectrometer was determined from the scattering of a Plexiglas to be 1.5-2.0 meV in full width at half maxima at 21.75 keV. The $Q$ resolution was ±0.48 nm$^{-1}$.

Metallic glass ribbons were synthesized by rapid quenching from the molten state using a single roller technique [5]. Two ribbons were stacked to be 30 μm in total thickness and located in a evacuated chamber with Capton X-ray windows. The measurement has been carried out at 25°C.

Before the inelastic experiments, static structure was confirmed in X-ray diffraction measurements utilizing BL04B2 beamline in SPring-8.

3. Results
Figure 1 shows dynamic structure factors, $S(Q,E)$, of CuZr$_2$ obtained from the measured scattering intensity after standard corrections of absorption and polarization, subtraction of Compton scattering and normalization by atomic form factor and so on. An inset in figure 1 shows static structure factors of CuZr$_2$ obtained from the diffraction experiment (full line) and estimated from the integrals of $S(Q,E)$ over energy-transfer within ±40 meV which is zeroth energy moment of $S(Q,E)$. The agreement of these two curves is good. One can see side bands with dispersion in both plus and minus $E$ sides of the central elastic contribution, which suggests existence of acoustic-like propagating wave in the metallic glass.

Figure 1. Dynamic structure factors, $S(Q,E)$, of metallic glass CuZr$_2$ at 25°C. In the inset, $S(Q)$ determined from the zeroth moment of the present $S(Q,E)$ is compared with $S(Q)$ obtained by X-ray diffraction.
4. Discussions

$S(Q,E)$ at each $Q$ value have been fitted using a model function as shown in figure 2. It is found that $S(Q,E)$ cannot be reproduced by a convolution of the resolution function and a summation of the central elastic contribution with delta function and the single excitation described by damped harmonic oscillator (DHO) model [3] corresponding to acoustic-like propagating wave (see the difference between the chained line and the experimental data in figure 2). Especially, experimental $S(Q,E)$ is rather intense than the fitted function in medium energy-transfer region between the central peak and the propagating excitation. Assuming that transverse mode with half excitation energy exists in this $E$ region, a model function with the central elastic peak and two excitations described by DHO model corresponding to longitudinal and transverse modes was applied. The fitting results was improved very much, however the residual function still exhibits two peaks in a low $E$ region of the both sides of the elastic peak. These peaks can be boson peaks that have been observed in many amorphous materials, although the peaked energies of almost constant about $\pm 1\text{meV}$ for every $Q$ values is too low compared with boson excitation since low energy-transfer sides of the transverse mode cover the region where the boson peak really lies.

Figure 3 shows the dispersion relations of longitudinal and transverse acoustic phonons with bars showing FWHM (full width at half maximum) of each DHO function. In the fitting, we assume that the sound velocity of the transverse mode is just half of that of the longitudinal mode, which in fact is only a good approximation at intermediate $Q$, but provides some first approximation to the data (further analysis is in progress). The dispersion curves drop on a sine curve in low $Q$ region like lattice dynamics of crystal. The sound velocity estimated from long-wavelength-limit is 4703 m/sec. The longitudinal dispersion curve suggests the existence of pseudo-Brillouin zone in the metallic glass.
5. Summary

IXS measurements of metallic glass CuZr$_2$ were performed and an acoustic-like propagating wave was observed. In addition of longitudinal mode, an additional mode that we take to be transverse was observed. Our fits also hint at the presence of a low-energy excitation similar to the boson peak can be distinguished at around ±2 meV.

Acknowledgements

This work was performed at the SPring-8 with the approval of the Japan Synchrotron Radiation Research Institute (JASRI) (Proposal No. 2006A1453 for the IXS experiments and No. 2006A1448 for the diffraction experiments).

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*Figure 3.* The dispersion relations of longitudinal and transverse modes in CuZr$_2$ are denoted by circles and triangles, respectively. Bars are shown in FWHM (full width at half maximum) of those modes analyzed using DHO (dumped harmonic oscillator) model function. The chained lines show sine curves fitted to the dispersion relations.