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The effect of composition on pressure-induced polyamorphism in metallic glasses

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ABSTRACT

Polymaorphism was recently revealed in metallic glasses (MGs) under high-pressure. The electronic structural evolution of solvent component was confirmed to be essentially responsible for the polyamorphic transition in MGs. In this paper, structural evolutions of $Ce_{68-x}La_xAl_{10}Cu_{20}Co_2$ (x = 0, 34, and 68) MGs were investigated by *in situ* high pressure X-ray diffraction measurements. The effect of composition on pressure-induced polyamorphism in MGs was discussed. The results indicated that polyamorphous in Ce-based MGs is strongly related to the Ce concentration. These results provide new insight for searching for polyamorphous and glass structure in MGs.

1. Introduction

Being a new class of disordered materials with many attractive properties, metallic glasses (MGs) have been extensively researched on their atomic structures and relationships to properties. Polymaorphism [1,2], in which multiple distinct amorphous states are formed from the same substance, has been extensively researched in a few non-metallic amorphous materials with open network structures, e.g., amorphous ice [3–4], chalcogenides [5], oxides [6], silicon [7], etc. These structural polyamorphic transitions from low-density amorphous state to high-density amorphous state often result from an increase in atomic coordination. Thus, the polyamorphic transition was thought to be impossible in MGs due to the nondirectional, densely packed atomic structure (coordination numbers up to 12–14) and the very low atomic mobility of these materials under hydrostatic high pressure [8]. Recently, polyamorphism was surprisingly observed in Ce-based MGs [9–15], in which the mechanism of polyamorphic transitions was revealed due to the pressure-induced the 4*f* electrons-delocalization of Ce atoms. The polyamorphic transitions between distinct amorphous states have also been reported in other rare earth element-based MGs (REMGs), suggesting that electronic structure inheritance of lanthanide-solvent atoms in REMGs [16–19]. However, what the physical and chemical controls behind polyamorphic transitions are still not clear. Answering this question will be an important guide for searching for more glass systems with shed new light on the investigation of glass structure. Changing Ce concentration will allow for a detailed study of the interplay between electronic and lattice role in the polyamorphic transitions. In this paper, a systematic investigations were carried out on $Ce_{68-x}La_xAl_{10}Cu_{20}Co_2$ (x = 0, 34, and 68) MGs using *in situ* high pressure X-ray diffraction (XRD). The effects of composition on polyamorphic transitions were discussed.

2. Experiments

The $Ce_{68-x}La_xAl_{10}Cu_{20}Co_2$ (x = 0, 34, and 68) MGs in at.% were prepared by arc-melting pure Ce, La, Cu, Al, and Co in a Ti-gettered argon atmosphere. Numerous details are found in Refs [20]. The *in situ* high pressure synchrotron XRD measurements with a focused X-ray beam of $26 \times 8\ \mu m^2$ at a wavelength of 0.6199 Å were carried out on 4W2 beamline at the Beijing Synchrotron Radiation Facility. More experimental details can be found in Refs. [17,18]. The atomic pair distribution function $g(r)$ were obtained by the program PDFgetX2 [21].

3. Results and discussion

Fig. 1 shows selected high pressure XRD patterns during compression for the $Ce_{68-x}La_xAl_{10}Cu_{20}Co_2$ (x = 0, 34, and 68) MGs at

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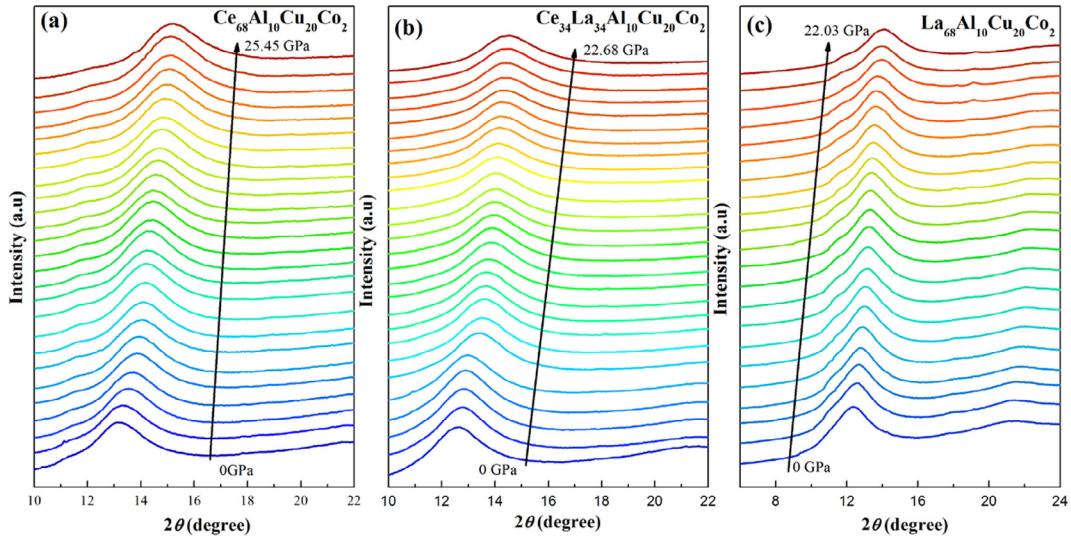

Fig. 1. *In situ* high pressure XRD patterns of $Ce_{68-x}La_xAl_{10}Cu_{20}Co_2$ (x = 0, 34, and 68) MGs under different pressures.

room temperature. The curve shows the broad diffraction maximum characteristic of amorphous structure and without any clearly visible diffraction peaks corresponding to crystalline phases. With increasing pressure, the main amorphous diffraction peak shifts to high 2θ, as expected for the densification effect of pressure. The reverse main amorphous diffraction peak position, $\lambda/2\theta$, correlates with the volume of glass having a power law function [22–24], which can be conveniently used to reflect the relative volume (density) change as a function of pressure. Fig. 2 shows the inverse main amorphous diffraction peak position, $\lambda/2\theta$, of the $Ce_{68-x}La_xAl_{10}Cu_{20}Co_2$ (x = 0, 34, and 68) MGs as a function of pressure during compression, which were estimated from the diffraction peak fitting using a Voigt line profile after subtracting baseline. It is found that at about 9 GPa, and 12.8 GPa, clear changes were detected for $Ce_{68}Al_{10}Cu_{20}Co_2$, and $Ce_{34}La_{34}Al_{10}Cu_{20}Co_2$ MGs, respectively. The changes indicate that the structure does exist differently between its initial configuration under high pressures. This confirmed that polyamorphic transitions indeed exist in

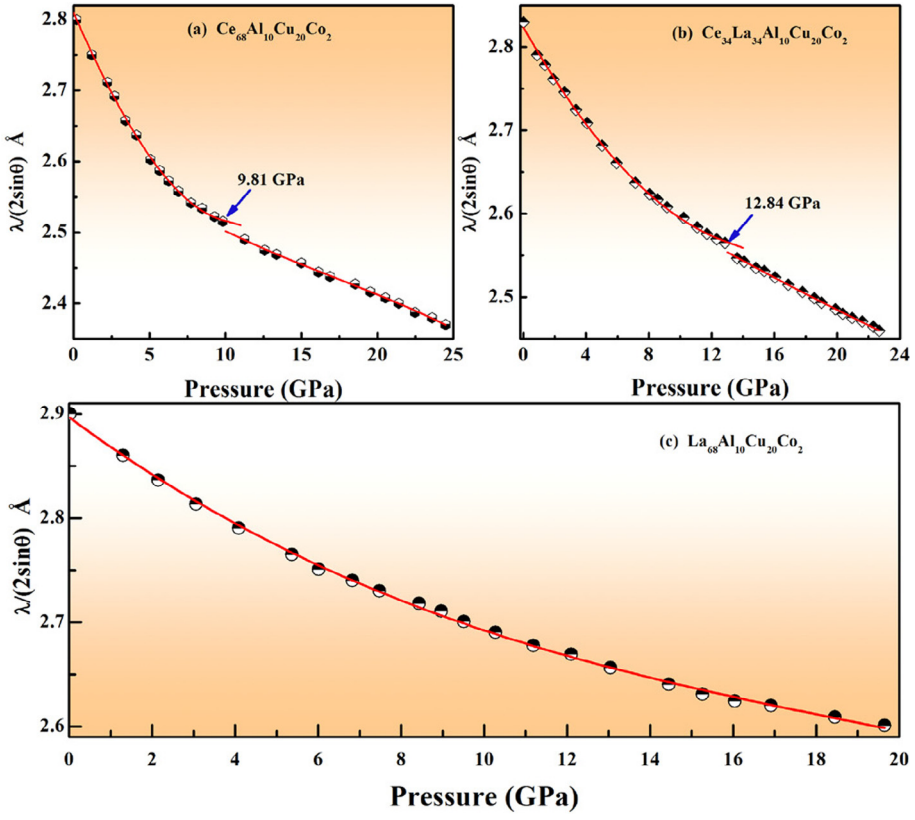

Fig. 2. The parameter ($\lambda/2\theta$) of the main amorphous peak, which is related to the reduced volume of the sample, as a function of pressure for $Ce_{68-x}La_xAl_{10}Cu_{20}Co_2$ (x = 0, 34, and 68) MGs.

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