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High-pressure x-ray diffraction of icosahedral Zr–Al–Ni–Cu–Ag quasicrystals

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The effect of pressure on the structural stability of icosahedral Zr–Al–Ni–Cu–Ag quasicrystals forming from a Zr65Al7.5Ni10Cu7.5Ag10 metallic glass with a supercooled liquid region of 44 K has been investigated by in situ high-pressure angle-dispersive x-ray powder diffraction at ambient temperature using synchrotron radiation. The icosahedral quasicrystal structure is retained up to the highest hydrostatic pressure used (approximately 28 GPa) and is reversible after decompression. The bulk modulus at zero pressure and its pressure derivative of the icosahedral Zr–Al–Ni–Cu–Ag quasicrystal are 99.10±1.26 GPa and 4.25±0.16, respectively. The compression behavior of different Bragg peaks is isotropic and the full width at half maximum of each peak remains almost unchanged during compression, indicating no anisotropic elasticity and no defects in the icosahedral Zr–Al–Ni–Cu–Ag quasicrystals induced by pressure.

Recently, after the discovery of the formation of icosahedral quasicrystals from Zr–Al–Cu–Ni metallic glasses,1 quasicrystals have been found by crystallization in many Zr-based alloy systems, such as ZrM (M=Pd and Pt),2–3 ZrNiM (M=Pd, Au, Pt, and Ti),4–5 ZrCuM (Al and Pd),6–7 ZrAlCuPd,7 ZrCuNiPd,7 ZrAlNiM (M=Cu, Pd, Au, and Pt),8–10 ZrAlNiCuM (M=Ti, Au, Pt, Pd, and Ag),7,11–17 and ZrTiCuNiBe.18 Inoue and co-workers15 further found that bulk quasicrystalline ZrAlNiCuM (M=Pd and Ag) alloys exhibit high strength and good ductility. The formation of quasicrystals in these alloys becomes very interesting. It has been demonstrated from several groups that Zr-based quasicrystals have a phase transformation into intermetallic compound(s) at high temperatures. In this work, we report the structural stability of the icosahedral Zr–Al–Ni–Cu–Ag quasicrystals forming from a Zr65Al17.5Ni10Cu7.5Ag10 metallic glass under hydrostatic pressure up to approximately 28 GPa by in situ high-pressure angle-dispersive x-ray powder diffraction (XRD) at ambient temperature using synchrotron radiation. It should be mentioned that stable quasicrystals against higher pressures on various conventional Al- and Ti-based systems have been reported.19–26 In most cases, the full width at half maximum (FWHM) of the Bragg peaks increases with pressure. It was suggested that the defects induced during compression stabilize the quasicrystalline structure. However, the results obtained here show that the quasicrystalline structure could be intrinsically stable during compression of about 17% volume contraction.

A ribbon sample of the Zr65Al17.5Ni10Cu7.5Ag10 metallic glass with a cross section of 0.03 mm×1 mm was prepared by the melt-spinning technique from a master alloy ingot prepared by arc melting in an Ar atmosphere. Thermal analysis of the as-prepared amorphous ribbon was performed in a differential scanning calorimeter (DSC) at a heating rate of 3 K/min under a flow of purified argon. The alloy exhibits an endothermic event characteristic of the glass transition Tg = 628 K, followed by two characteristic exothermic events Tx1 = 672 K and Tx2 = 730 K, and 34.1 and 40.4 J/g, respectively, indicating two-stage amorphous-to-quasicrystalline and quasicrystalline-to-intermetallic phase transformation processes, which are consistent with data reported in the literature for the alloy.11–17 Subsequent annealing for preparation of Zr-based quasicrystals was performed on the as-quenched amorphous ribbon in a vacuum of 1×10–6 Torr at 663 K for 2 h. The icosahedral quasicrystalline structure of the annealed sample was confirmed by x-ray powder diffraction and transmission electron microscopy. The average grain size of the icosahedral quasicrystals was approximately 45 nm. A flake (approximately 20 μm) of the annealed sample was loaded into a diamond-anvil cell with a Re gasket of 150 μm in hole. In order to insure the hydrostatic conditions up to 30 GPa, liquid He (2000 atm) was used as the pressure transmitting medium. The actual pressure was calculated from the wavelength shift of the ruby line using the non-linear pressure scale of Mao et al.27 In situ high-pressure angle-dispersive x-ray powder diffraction measurements of the annealed sample were performed at the BL10XU beamline, SPring8, Japan, with a wavelength of 0.49592 Å. The diffraction patterns were recorded using an image plate, which provides complete information in the form of a possible preferential orientation of diffracting grains and a better sample averaging by integrating the Debye–Sherrer rings by means of a two-dimensional data analysis program.

A large number of XRD patterns were recorded at pressures ranging from 0 to approximately 28 GPa at ambient temperature. In the annealed sample, the two-dimensional diffraction patterns recorded did not show any preferential

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The quasilattice constant at ambient pressure is found to be $q$, which is the quasilattice constant. The quasilattice constant at ambient pressure is found at $q_{\text{ambient}}$. Higher two theta angles give three Bragg peaks that shift monotonously to the right with increasing pressure. The Bragg peaks do not disappear, nor does any new peak appear, so that the alloy remains icosahedral. After release of pressure (from 28 GPa), Bragg peaks recover their initial positions. The relative intensity and FWHM of the Bragg peaks are almost unchanged within experimental uncertainty. This infers no distinguishable anisotropies in the icosahedral Zr–Al–Ni–Cu–Ag quasicrystals.

A question of particular interest is whether the sample exhibits any anisotropic elasticity. To address this question, we have plotted the compressibility in our case $V(P)/V(0)$ of the icosahedral Zr–Al–Ni–Cu–Ag quasicrystals using the Birch–Murnaghan equation.

Table I lists the zero-pressure bulk modulus ($B_0$) and its pressure derivative ($B'_0$) for the Bragg peaks as well as the averaged values of the three peaks for the icosahedral Zr–Al–Ni–Cu–Ag quasicrystals using the Birch–Murnaghan equation. Figure 4 plots compression data obtained by averaging the three Bragg peaks. It is clear that the compressibility is equivalent in all directions within experimental uncertainty.

In conclusion, the effect of hydrostatic pressure on the icosahedral Zr–Al–Ni–Cu–Ag quasicrystals is intrinsically stable up to 28 GPa; (2) no defect, dislocation, and phason were induced during compression; and (3) no preferential orientation of any symmetry lines was detected during compression. This indicates that the sample exhibits anisotropic elasticity.

![FIG. 2. Pressure dependence of d spacing for three Bragg peaks of the icosahedral Zr–Al–Ni–Cu–Ag quasicrystals.](image)

![FIG. 3. Pressure dependence of the FWHM for three Bragg peaks of the icosahedral Zr–Al–Ni–Cu–Ag quasicrystals. FWHM of the [110000] and [101000] peaks are added with 0.1° and 0.25°, respectively.](image)

**TABLE I.** Zero-pressure bulk modulus ($B_0$) and its pressure derivative ($B'_0$) for the Bragg peaks as well as the averaged values of the three peaks for the icosahedral Zr–Al–Ni–Cu–Ag quasicrystals using the Birch–Murnaghan equation, together with data reported in the literature.

<table>
<thead>
<tr>
<th>$B_0$ (GPa)</th>
<th>$B'_0$ (GPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100000</td>
<td>100.04 ± 1.19</td>
</tr>
<tr>
<td>110000</td>
<td>98.04 ± 1.21</td>
</tr>
<tr>
<td>101000</td>
<td>99.25 ± 1.52</td>
</tr>
<tr>
<td>Average</td>
<td>99.10 ± 1.26</td>
</tr>
<tr>
<td>Al–Cu–Fe$^a$</td>
<td>155</td>
</tr>
<tr>
<td>Al–Pd–Mn$^b$</td>
<td>128</td>
</tr>
<tr>
<td>Al–Cu–Ru$^c$</td>
<td>128</td>
</tr>
<tr>
<td>Al–Pd–Re$^c$</td>
<td>180</td>
</tr>
<tr>
<td>Al–Ni–Co$^d$</td>
<td>120</td>
</tr>
<tr>
<td>Al–Pd–Mn$^e$</td>
<td>100 ± 12</td>
</tr>
<tr>
<td>Ti–Zr–Ni$^f$</td>
<td>130 ± 10</td>
</tr>
<tr>
<td>Ti–Zr–Ni–H$^g$</td>
<td>105 ± 10</td>
</tr>
<tr>
<td>Ti–Zr–Ni$^h$</td>
<td>173 ± 5</td>
</tr>
</tbody>
</table>

$^a$Reference 20.  
$^b$Reference 21.  
$^c$Reference 22.  
$^d$Reference 23.  
$^e$Reference 24.  
$^f$Reference 25.  
$^g$Reference 26.  
$^h$Reference 26.
approximately 28 GPa) on the structural stability of the
icosahedral Zr–Al–Ni–Cu–Ag quasicrystals forming from a
Zr$_{65}$Al$_{7.5}$Ni$_{10}$Cu$_{7.5}$Ag$_{10}$ metallic glass has been investigated
in situ using synchrotron radiation. It is found that
the icosahedral quasicrystalline structure in the sample is in-
trinsically stable up to 28 GPa. The bulk modulus at zero
pressure and its pressure derivative of the icosahedral
Zr–Al–Ni–Cu–Ag quasicrystals is 99.10±1.26 GPa and 4.25
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