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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 Zr_{65}Al_{7.5}Ni_{10}Cu_{7.5}Ag_{10} 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. © 2001 American Institute of Physics.

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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),11,12–17 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 compounds at high temperatures. In this work, we report 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 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 Zr_{65}Al_{7.5}Ni_{10}Cu_{7.5}Ag_{10} 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 \( T_g = 628 \text{ K} \), followed by two characteristic exothermic events \( T_x = 672 \text{ K} \) and \( T_x = 730 \text{ K} \), 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 \( 1 \times 10^{-6} \text{ Torr} \) 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 of 45 nm. A flake (approximately 20 \( \mu \text{m} \)) of the annealed sample was loaded into a diamond-anvil cell with a Re gasket of 150 \( \mu \text{m} \) 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 nonlinear 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–Scherrer 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...
pressed by q

ambient pressure is found to be q~ the quasilattice constant. The quasilattice constant at am-

~ golden mean, 1.618. As an example, the q~ found at higher two theta angles lattice vector. The Bragg peaks shift monotonously to the 3 , for the three most distinguishable Bragg peaks (100000, 110000, 101000) as a function of pressure. 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.\[28\] 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. This infers no distinguishable anisotropies in the icosahedral Zr–Al–Ni–Cu–Ag quasicrystals.

In conclusion, the effect of hydrostatic pressure (up to 28 GPa) is found to be q~ the quasilattice constant. The quasilattice constant at ambient pressure is found to be q~ the golden mean, 1.618. As an example, the q~ found at higher two theta angles lattice vector. The Bragg peaks shift monotonously to the 3 , for the three most distinguishable Bragg peaks (100000, 110000, 101000) as a function of pressure. 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.\[28\] 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. This infers no distinguishable anisotropies in the icosahedral Zr–Al–Ni–Cu–Ag quasicrystals.

<table>
<thead>
<tr>
<th>B_0 (GPa)</th>
<th>B_0'</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 [f]</td>
<td>100±12</td>
</tr>
<tr>
<td>Ti–Zr–Ni [f]</td>
<td>130±10</td>
</tr>
<tr>
<td>Ti–Zr–Ni–H [f]</td>
<td>105±10</td>
</tr>
<tr>
<td>Ti–Zr–N [f]</td>
<td>173±5</td>
</tr>
</tbody>
</table>

\[a\] Reference 20. \[b\] Reference 21. \[c\] Reference 22. \[d\] Reference 23. \[f\] Reference 24.
Zr$_{65}$ Al$_{7.5}$ Ni$_{10}$ Cu$_{7.5}$ Ag$_{10}$ metallic glass has been investigated, and the icosahedral Zr–Al–Ni–Cu–Ag quasicrystals forming from a Al–Ni–Cu–Ag quasicrystal is intrinsically stable up to 28 GPa. The bulk modulus at zero pressure of the icosahedral quasicrystalline structure in the sample is approximately 28 GPa. 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.

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