Electronic Structure and Glass Forming Ability in Bulk Metallic Glasses

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Bulk metallic glass (BMG) is a class of non-crystalline metallic system with some outstanding physical and chemical properties. BMGs are significantly different from crystalline materials due to their lack of long-range order (LRO) and grain boundaries. Therefore, dislocations, which mediate plasticity in crystalline materials, cannot operate. Consequently, BMGs are usually extremely strong with a maximum elastic strain of about 2.3 % and a yield point approaching the theoretical strength limit [1,2]. The ideal BMG for structural applications should have both excellent glass forming ability (GFA) that avoids crystallization, and intrinsic ductility that minimizes brittle fracture. A computational-based approach to rationally search and evaluate both the GFA and the intrinsic ductility of BMGs through atomistic simulation and high-fidelity density functional calculations offers a realistic hope in reaching this goal.

There has not been much recent effort in using first-principles methods to study the electronic structure of BMGs. The main difficulties are: (1) the non-crystalline nature of BMGs, and (2) the lack of highly accurate and sufficiently large structure models to be used for the evaluation of their physical properties. Detailed information on the atomic-scale interactions and their relation to the short range order (SRO) and medium range order (MRO) are missing. Most current research focuses on geometric description of structural units in BMG and their implications on GFA.

In this talk, I will present some preliminary results on large-scale *ab initio* calculations of the electronic structure in some BMG models to elucidate the atomic scale understating of the structures of BMG. Fig. 1 shows a supercell model of a binary $Zr_{50}Cu_{50}$ BMG with 1,024 atoms. The model was first generated using classical molecular dynamics (MD) after long annealing steps and then relaxed with VASP with high precision. The fully-relaxed structure is then used as input for the electronic structure calculation using OLCAO method [3]. We have obtained three such BMG models: $Zr_{50}Cu_{50}$, $Zr_{48.5}Cu_{51.5}$, and $Zr_{50}Cu_{40}Al_{10}$. The calculated DOS for these models are shown in Fig. 2. The result appears to support the notion that the stability of the glass (hence the GFA) correlates with a minimum in the DOS near the Fermi level [4], and is consistent with experimental observations [5, 6]. The calculation also shows that there is an average charge transfer of 0.683 electrons from Zr to Cu in the $Zr_{50}Cu_{50}$ model. It is possible to resolve the DOS into atomic components, or PDOS. By recombining the 1024 PDOS into groups of atoms according to their local geometric configurations (icosahedra, Kasper polyhedra etc.), we would be able to make a direct connection between electronic structure and local geometric structure in BMG. The elastic constants C_{ij} and the mechanical properties of BMG model can also be calculated. From the C_{ij}, the bulk modulus (K), shear modulus (G), Young's

modulus (E), and Poisson's ratio (ν) can be evaluated. Recent discovery indicated an empirical correlation between the fracture energy and Poisson's ratio for MGs and network glasses [7]. Amorphous materials with high Poisson's ratios are ductile whereas those with low Poisson's ratios are brittle. These and other results will be presented and discussed.

References:

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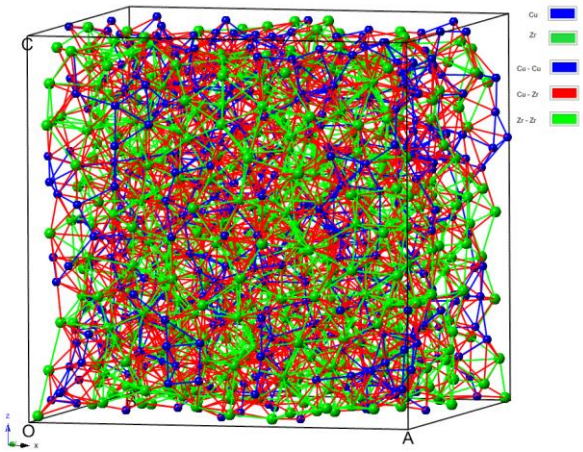

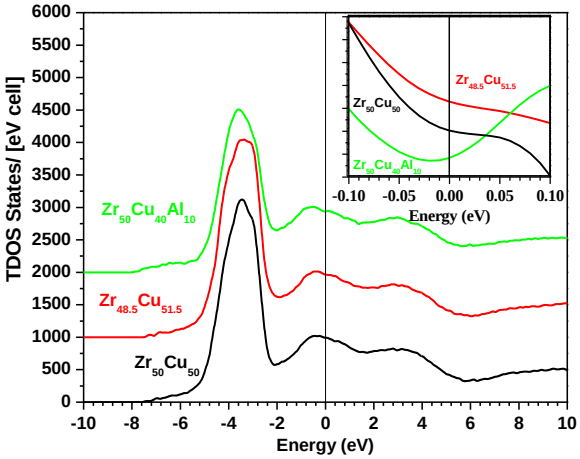

FIG.1 (left) Sketch of a 1024 atom supercell model for $Zr_{50}Cu_{50}$.

FIG.2 (right) DOS of three metallic glass models: $Zr_{50}Cu_{50}$ (black), $Zr_{48.5}Cu_{51.5}$ (red) and $Zr_{50}Cu_{40}Al_{10}$ (green). The vertical line is the Fermi level. The inset shows the DOS within a narrow region of E_F. The slopes and the shape near E_F are correlated with the relative stability of the glass.