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A study of lattice elasticity from low entropy metals to medium and high entropy alloys

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An equal-molar CoCrFeMnNi, face-centered-cubic high-entropy alloy system and a face-centered-cubic stainless steel described as a medium-entropy system, are measured by *in situ* neutron-diffraction experiments subjected to continuous tension at room and several elevated temperatures, respectively. With spallation neutron, the evolution of multiple diffraction peaks is collected simultaneously for lattice-elasticity study. Temperature variation of elastic stiffness of a single face-centered-cubic-phase Ni and a single face-centered-cubic-phase Fe are compared as low-entropy metals. The CoCrFeMnNi high-entropy alloy shows distinct lattice anisotropy.

Keywords: Neutron diffraction; Metal and alloys; Elastic behavior; Microstructure; High entropy alloy

In 2014, Gludovatz et al. [1] report superior fracture-resistant property of an equiatomic, face-centered-cubic, CoCrFeMnNi-high-entropy alloy (HEA) for cryogenic applications. Moreover, several nontrivial mechanical behavior of this kind of HEA is observed by Yeh et al. [2], Nieh et al. [3] and George et al. [4], respectively. Besides its practical application [1], the underneath mechanical mechanisms of this CoCrFeMnNi-high-entropy alloy are unclear. For example, how equiatomic-element mismatch affect its bonding [5]? Would Vegard's law be valid [6]?

For metallic systems, numerous works have demonstrated the importance of ordering for compositions [7,8]. Comparing with the metallic systems, Ashyby's summarizes the basic mechanical and thermal properties of materials and reveals the sub-range associated with each material class [9]. Generally, the energy-driven moduli of the soft materials are smaller than their counterparts. The moduli of soft materials typically show temperature-dependent entropy-driven and energy-driven elastic behavior. For the configuration ordering, the entropic elasticity originates from the thermodynamics. When there is configuration rearrangement, the enthalpy of phase transition can well quantify the level of entropic effect after Clausius–Clapeyron relation [10–13]. For the conformation ordering, spatial distribution of a cluster of atoms or molecules brings significant entropy-driven elasticity to local potential energy, too. [14–18] For the metallic systems, such temperature-dependent moduli subjected to order–disorder transition have also been quantified [19,20]. For long-range-order–disorder transition, a step-change in Young's modulus of the Cu_3Au system is observed at critical temperature [19,20]. Besides the aforementioned long-range-order–disorder transition, there is deformation-induced entropic effect on the modulus found in bulk-metallic-glass (BMG) system [21]. Falk and Langer postulate the dynamics of two-state shear transformation which irreversible motions are governed by local entropic fluctuations in the volumes of the transformation zones at temperatures far below the glass transition. Above all, several results have reported the entropic effect on the elasticity of the metallic systems in terms of the long-range-order–disorder transition [19,20], martensitic transformation [22], viscoplasticity [21], shear transformation [21], and other plasticity-related behavior [23].

Finally, Hufnagel et al.'s *in situ* synchrotron diffraction measurements reveal entropic contribution for the stiffness of metallic glasses within elastic limit [24]. Wagner et al.'s molecular dynamics simulation show local elastic properties of a metallic glass [25]. Their results show that the local potential energy can vary significantly in space for non-crystalline materials [24,25]. However, the BMGs' local modulus distribution is very different from the crystalline

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with a long-range order. For long-range order existing in space, a unified elastic modulus is expected [25]. Based on such a fact, for polycrystalline metals, the Taylor theory assumes that the strain tensor is symmetrical [26]. Similarly, Molinari et al. use a mean field approach to describe the deformation of polycrystals of an ellipsoidal inclusion embedded in a homogeneous matrix with a unified elastic modulus [27].

In this paper, we focus on *hkl*-dependent elasticity that is the consequence originated from the aforementioned effects. Explicitly, it is an extension of previous reports [4,28] which specifies a range of temperature-dependent mechanical-behavior transitions. Otto et al. found strong temperature-dependent behavior on the tensile properties of this system ranging from 77 K to 1073 K [4]. Specifically, Gali and George discovered that the transition between the thermal and athermal regions of the HEAs is higher than that of typical binary alloys [28]. Yeh et al. [2] have concluded such a behavior could reflect the severe distortion in the high-entropy alloys, which creates much more resistance than traditional alloys at the same temperature [29]. The extended X-ray absorption fine structure evidence shows that the lattices of HEAs are distorted by the heterogeneity of local chemical distributions [30]. The aforementioned recent research lead us to ask: could this local compositional heterogeneity of the equiatomic high-entropy alloy be sufficient enough to break tensor symmetry and collaborate with energy-driven elasticity?

Here we show the equiatomic, face-centered-cubic CoCrFeMnNi metallic system as a high-entropy alloy (HEA); a face-centered-cubic 304L stainless steel (with a weight percentage of C:0.022; Cr:18.10; Ni:8.040; Mn:1.390; Mo:0.550; Cu:0.465; Si:0.256; Co:0.144; N:0.080; W:0.060; V:0.060; P:0.039; S:0.027; Sn:0.015 and balance Fe) as a medium-entropy alloy (MEA), and pure Fe and pure Ni as low-entropy metals (LEMs). We facilitate the advantage of the *in situ* neutron diffraction measurements for its great gauge volume to investigate the ensemble-averaged lattice elasticity of the HEA and the MEA to compare with that of the LEMs. The procedures of *in situ* diffraction
Q3 experiments are described in the section of Methods, which is similar to Wu et al.'s room-temperature measurement [31]. The difference is that besides room temperature experiments, we measure both the HEA and MEA at several elevated temperatures as shown in the Supplementary Information. With the event-based data acquisition at Spallation Neutron Source (SNS) of the Oak Ridge National Laboratory (ORNL), neutron diffraction patterns were collected under continuously-loading mode. The feature is especially useful for the current investigation to observe dynamics loading condition [32]. Moreover, the advantage of the spallation-neutron-source materials-engineering diffractometer, such as VULCAN [32] of SNS, is that multiple diffraction peaks are collected simultaneously.

We follow Wu et al.'s presentation [31] to plot elastic modulus of different *hkl* planes as a function of anisotropy factor (A_{hkl}) in Fig. 1(a) for pure Ni, 1(b) for pure Fe, 1(c) for 304L stainless steel, and 1(d) for CoCrFeMnNi HEA, respectively. The definition of A_{hkl} is shown below.

$$A_{hkl} = \frac{h^2k^2 + k^2l^2 + h^2l^2}{(h^2 + k^2 + l^2)}$$

Fig. 1(a) and (b) is based on Ledbetter and Reed's temperature-dependent elastic-stiffness-matrix data [33]. We calculate the mechanical elastic constants for its face-centered-cubic structure with Kröner model [34,35]. Fig. 1(c) and (d) shows our experimental results. Measurements at different temperatures from room temperature to elevated temperatures are labeled in the insets of Fig. 1(a)–(d) accordingly.

Generally, in Fig. 1(a) and (b), the moduli and the anisotropy factor (A_{hkl}) exhibit a linear relationship. Hutchings et al. [36] have summarized several metallic systems showing the same linearity for their homogenous average elastic properties of the aggregate. Within elastic-deformation limit, there are no plasticity-induced heterogeneous-intergranular stresses. In Fig. 1(a) and (b), pure Ni and pure Fe show the same feature because their grains are considered chemically and mechanically uniform throughout the aggregate as Taylor [26], Sachs [37], and Kröner [38] constitute.

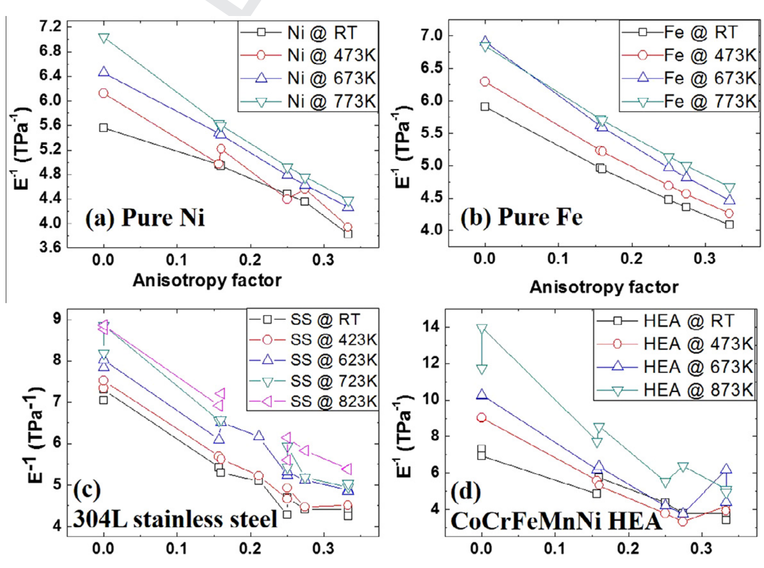

Figure 1. Variation of reciprocal moduli subjected to anisotropy factor for (a) pure Ni, (b) pure Fe, (c) 304L stainless steel, and (d) CoCrFeMnNi HEA.