Anomalous random correlations of force constants on the lattice dynamical properties of disordered Au-Fe alloys

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Gold iron (Au-Fe) alloys are of immense interest due to their biocompatibility, anomalous Hall conductivity, and applications in various medical treatments. However, irrespective of the method of preparation, they often exhibit a high level of disorder with properties sensitive to the thermal or magnetic annealing temperatures. We calculate the lattice dynamical properties of $Au_{1-x}Fe_x$ alloys using density functional theory methods where, being multisite properties, reliable interatomic force constant (IFC) calculations in disordered alloys remain a challenge. We follow a twofold approach: (1) an accurate IFC calculation in an environment with nominally zero chemical pair correlations to mimic the homogeneously disordered alloy and (2) a configurational averaging for the desired phonon properties (e.g., dispersion, density of states, and entropy). We find an anomalous change in the IFC's and phonon dispersion (split bands) near x = 0.19, which is attributed to the local stiffening of the Au-Au bonds when Au is in the vicinity of Fe. Other results based on mechanical and thermophysical properties reflect a similar anomaly: Phonon entropy, e.g., becomes negative below x = 0.19, suggesting a tendency for chemical unmixing, reflecting the onset of a miscibility gap in the phase diagram. Our results match fairly well with reported data wherever available.

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Gold (Au) and iron (Fe) and their alloys continue to attract attention. Due to the higher magnetic state of Fe in Au-Fe than in pure Fe, various properties have been studied, including thickness-dependent spin-glass behavior and anomalous Hall conductivity in Fe/Au multilayers [1–5]. Due to their exceptional biocompatibility and favorable physical properties, Au-Fe nanoparticles find various applications in medical sciences [6-10]. Gold-rich Au-Fe alloys form a face-centeredcubic (fcc) structure. Although fcc is a high-temperature phase, Au-Fe alloys up to 53 at. % Fe are reported to be stabilized easily at room temperature [11-14]. Due to the sensitivity of magnetic and chemical properties to annealing temperatures, these alloys require the most care in their synthesis, especially as disorder is quite common and difficult to control. Hence, chemical disorder plays an important role in their anomalous structural and magnetic properties.

For alloys to have useful applications, mechanical stability is a necessary criterion. Studying the lattice dynamics provides direct stability information and gives an idea about the local atomic environment and related phenomena. Experimentally, techniques, such as nuclear resonant inelastic x-ray scattering, inelastic neutron scattering, and Mossbauer spectrometry are used to investigate the elementary excitation in disordered alloys [14,15]. But, a reliable *ab-initio* theoretical approach to properly address the lattice dynamics of disordered alloys is lacking, mainly due to the requirement of an appropriate scheme to perform configurational averaging and the evaluation of random interatomic force constants. In fact, the challenge is to address the off-diagonal disorder arising out of the force constant matrix between two sites. In addition, the sum rule obeyed by the force constants implicitly makes the disorder at a site dependent upon its neighborhood, i.e., environmental disorder.

Historically, various approximate models are proposed to address disorder. The virtual crystal approximation (VCA) [16] and the coherent potential approximation (CPA) are two widely known single-site examples, which also suffer from deficiencies [17-21]. VCA, the simplest among many, involves simple compositional averages of the constituent potentials and completely ignores environmental effect. The single-site CPA suffers from capturing the multisite effects expected in lattice dynamics, such as off-diagonal and environmental disorders. Some generalizations for the CPA, e.g., itinerant CPA [21], dynamical cluster approximation [22], and its first-principles version (i.e., nonlocal CPA [19,20]) address the two-site disorder. These methods consist of various promising features, but they usually are limited to specific types of off-diagonal disorder or to small clusters due to computational expense. The special quasirandom structure (SQS) technique [23] is being utilized more often to estimate environmental effects of disorder. It involves a fully ordered cell (useful for the band-structure method) in a layered arrangement of atoms that nominally exhibits zero chemical pair correlations (within a specified range of neighboring two to three shells) and mimics those of the homogeneously disordered alloy. To predict the lattice dynamical properties of disordered systems, an accurate calculation of force constants as well as an appropriate configurational average over the disorder environment are equally important.

Here, we combine two techniques to correctly address the above issues: the SQS and augmented space recursion (ASR). ASR is a powerful method to capture multisite disorder effects as required in the phonon problem. A detailed description of the ASR formalism for phonons is given in the Supplemental Material (Sec. S1) [24]. For a given size and symmetry cell,

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TABLE I. Force constants (N/m) for $Au_{1-x}Fe_x$ along [110]. The measured data for pure Au (x = 0) are 16.63, 20.82, and -8.62 along the 110_{xx} , 110_{xy} , and 110_{zz} directions, respectively [14].

x	1.00	0.50	0.25	0.19	0.06	0.00	Direction
Au-Au Fe-Fe Au-Fe	9.29	26.39 14.08 13.89	21.79 9.16 10.98	19.05 2.39 10.98	16.66 9.38 9.20	17.52	110_{xx} 110_{xx} 110_{xx}
Au-Au Fe-Fe Au-Fe	17.88	30.57 14.48 16.78	26.18 1.49 12.05	23.06 2.92 13.20	20.33 10.67 10.85	21.03	110_{xy} 110_{xy} 110_{xy}
Au-Au Fe-Fe Au-Fe	8.15	-6.37 2.68 -1.62	-6.95 -8.01 -2.86	-6.64 -2.35 -3.17	-6.52 -1.60 -2.84	-5.94	$ \begin{array}{r} 110_{zz} \\ 110_{zz} \\ 110_{zz} \\ 110_{zz} \end{array} $

the SQS is used in conjunction with the small displacement method [25] to calculate the estimated force constants in a disordered alloy. ASR then performs the configurational averaging with these disordered force constants. We calculated the phonon dispersion, density of states, lifetime, vibrational entropy, and thermomechanical properties for $Au_{1-x}Fe_x$.

Spin-polarized density functional theory [26] calculations are performed with a projected augmented wave [27] basis using the local-density approximation as implemented in the Vienna ab initio simulation package [28,29]. We chose an optimal 32-atom SQS unit cell [30] to perform all the calculations for $Au_{1-x}Fe_x$ for x = 0.50, 0.25, 0.19, and 0.06, providing good accuracy. For the SQS structure of a given alloy, all atoms were relaxed to achieve energy (force) convergence of up to 10^{-6} eV (10^{-3} eV/Å). We use a high-energy cutoff of 450 eV with a Monkhorst-Pack $6 \times 6 \times 6$ k-mesh grid [31]. For x =0.50, 0.25, 0.19, and 0.06, respectively, the optimized lattice parameters in Au_{1-x}Fe_x were 3.83, 3.96, 4.00, and 4.05 Å. The experimental lattice parameters for x = 0.50, 0.30, 0.20, and0.03 are 3.908, 3.991, 4.026, and 4.072, respectively, which compared fairly well with theory. Phonons were calculated using the small displacement method as implemented in PHON [25], and the atomic force fields were obtained using 48, 96, 96, and 19 displacements for the respective x's. For elastic constants, we used the PBEsol exchange-correlation functional [32] with a $10 \times 10 \times 10 \Gamma$ -centered k mesh for total-energy calculation at different strains.

As the SQS provides structures with reduced symmetry (not fcc), the force constant matrix becomes random and asymmetric, which cannot be used directly in ASR for configurational averaging. To extract meaningful parameters for the proper fcc symmetry, a directional average mapping method is adopted. For fcc symmetry, we mapped all 12 nearest-neighbor matrix elements for Au-Au, Fe-Fe, and Au-Fe pairs in Au_{1-x}Fe_x at each site along [110] using $\phi^{101} = T^{\dagger}\phi^{110}T$, where *T* is the transformation matrix along the different directions.

The averaged force constants for all pairs are tabulated in Table I. Notably, Au-Fe force constants become stiffer as we increase the iron concentration (% Fe). The addition of Fe in pure Au makes the Au-Au pair stiffer, but Fe-Fe either becomes softer or remains unaffected. Interestingly, $Au_{81}Fe_{19}$ shows a turning point where the force constant matrix elements exhibit a nonmonotonous change. This anomaly also is reflected in the phonon dispersion, entropy, and other properties. The origin of this cannot be explained simply by the changes in lattice

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parameters or the overall electron DOS at the Fermi energy (E_F) . Below we provide a deeper explanation.

Au-Fe alloys are known for their rich magnetic properties [11,37]. Pure Fe in its stable bcc phase has a magnetic moment of 2.13 μ_B /atom, in agreement with previous theoretical and experimental data [38,39]. As % Fe decreases from 0.50 to 0.06, the Fe moments increase from 2.71 to 2.99 μ_B /atom also found previously [14,40]. fcc Au is a nonmagnetic metal. However, we found that the Au 5*d* moments are 0.083 (0.146) in Au₇₅Fe₂₅ (Au₅₀Fe₅₀), similar to other reported theories and experimental values of 0.099 (0.197), respectively [11]. (The detailed result can be found in the Supplemental Material [24] Sec. S2.)

Figure 1 shows phonon dispersions for $Au_{1-x}Fe_x$ along high-symmetry directions. Note the split band behavior with $x \ge 19\%$. Such splittings normally arise for systems with dominant masses or force constant disorders. Ni-Pt is a classic example of such behavior. In their elemental phase, Pt-Pt force constants are 55% larger than Ni-Ni. Although the force constant difference here is not that significant, the mass difference is higher $(M_{\rm Au}/M_{\rm Fe} = 3.53)$. Such splitting is a consequence of strong resonance, arising mainly out of dominant mass disorder here. Near resonances the FWHM become very large as is clear in Fig. 1. ASR is expected to correctly address both mass and force constant disorders, mainly in the higher-frequency region, as demonstrated in our earlier papers [41,42]. Figure 1 also shows the phonon DOS where the higher- (lower-) frequency region is dominated by Fe (Au) as expected by mass. It also explains the increase in the number of states in the higher-frequency region as the % Fe increases. Our calculated phonon dispersion and DOS compare fairly well with previous experimental data [14]. The anomalous band splitting arises for $x \ge 0.19$, the turning point in the force constants (Table I). This behavior can be understood from the evolving nature of the Au-Au bond in an Fe matrix. When Fe is substituted in Au, there are two types of force constants that Au-Au pairs acquire. The pairs that do not contain Fe in their vicinity have force constants similar to that of pure Au. However, pairs that exist in the neighborhood of Fe increasingly stiffen as % Fe increases, which causes an increase in the energy of some Au modes above the cutoff energy of the Au modes and hence causes the splitting. This behavior can be found explicitly in thermomechanical properties of the alloy as seen below.

For a material, thermomechanical parameters are related directly to the second-order elastic constants. For a cubic crystal, there are three independent elastic constants denoted by C_{11} , C_{12} , and C_{44} . Here we use a strain-energy approach [30,43] to evaluate C_{ii} at various % Fe, along with the bulk modulus (B), the shear moduli $(G_V, G_R, \text{ and } G_H)$, Young's modulus (Y), the shear constant (C'), the Cauchy pressure (C_P) , Poisson's ratio (v), the Kleinman parameter (K_{ζ}) , Zener's anisotropy ratio (A^Z) , Pugh's indicator (P), Lames's coefficients (λ and μ), the longitudinal and transverse sound wave velocities $(v_l \text{ and } v_t)$, the Debye temperature (Θ_D) , the high-temperature limit to the thermal conductivity as obtained via Clarke's model ($\kappa_{\min}^{\text{Clarke}}$) [44], and Cahill's model $(\kappa_{\min}^{\text{Cahill}})$ [45]. All the properties help assess the mechanical stability of the material (see the Supplemental Material [24] (Secs. S3 and S4) for details).



FIG. 1. (Left) Using force constants from the 32-atom SQS and phonon dispersion for Au_{1–x}Fe_x along high-symmetry [ζ 00], [ζ ζ 0], and [ζ ζ ζ], where $\zeta = |\vec{k}|/|\vec{k}_{max}|$ for reciprocal-space vector \vec{k} . Longitudinal (*L*) and transverse (*T*) modes are indicated. The bars indicate the calculated full width at half maxima (FWHM). (Right) Projected density of states (DOS).

Table II presents the calculated values of these quantities along with available experimental data. Our data $(C_{ij}$'s) agree

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within 2–5% of the experiment for Au [33]. For pure Fe, calculated C_{11} is overestimated by 25%, whereas C_{12} and C_{44} are estimated by 19% and 11%, respectively, compared to the experiment. This is due, as is well known, to the generalized gradient approximation exchange-correlation function used here, which tends to underestimate the lattice constants for 3*d* transition metals [46,47]. The mechanical stability criteria of Born-Huang [48] ($C_{11} - C_{12} > 0$, $C_{11} + 2C_{12} > 0$, and $C_{44} > 0$) are satisfied for both the elements and the alloys. The calculated C' = 20.5 GPa for Au₈₁Fe₁₉ compares well with that measured at 20.7 GPa [34]. All our calculated results are expected to be within 10–15% of the measured values and will be interesting to be verified experimentally.

Pure Au has high B but small G_H , which makes it very ductile, as seen by its indicator from Pugh (5.92) [49] where materials with P > 1.75 are ductile. With increasing % Fe, the size and coupling force mismatch make the system stiffer (see Table I), resulting in reduced ductility and a higher Young's modulus. $C_p > 0$ [50] suggests metallic bonding character as well as higher conductivity. High- ν values confirm this. $A^Z >$ 1 [51] for both the elements and the alloys points to highly anisotropic deformation in the material (a higher possibility of microcracks). K_{ζ} [52] (between 0 and 1) indicates the nature of the bonding. A lower (higher) K_{ζ} indicates dominant bond bending (stretching) as found here for Au-Fe. Lame's constant λ suggests large incompressibility of these alloys. Debye temperature (Θ_D) increases with disorder, attributed to higher mass fluctuation and an increase in the frequency of the thermal vibrational modes.

At x = 0.19, a similar anomaly (as in force constants and phonon dispersion) is encountered in some elastic properties (e.g., C_{11} , C_{22} , B, and C_P), reflecting the dominant force constant disorder and the emergence of the resonance mode in the dispersion. Such unusual behavior also is predicted by Munoz *et al.* [14] at x = 0.2, which is attributed to the increasing stiffness of the Au-Au bonds with increasing % Fe, and primarily a local effect.

Next, we have calculated the temperature dependence of excess vibrational entropy $\Delta S_{\text{vib}} = (1 - x)\Delta S_{\text{vib}}^{\text{Au}} + x \Delta S_{\text{vib}}^{\text{Fe}}$, where $\Delta S_{\text{vib}}^{\text{Au}} (\Delta S_{\text{vib}}^{\text{Fe}})$ are the partial contributions to vibrational entropy from Au (Fe), respectively, at each *x*. These are calculated as

 $\Delta S_{\text{vib}}^{M} = S_{\text{vib}}^{M}(\text{alloy}) - S_{\text{vib}}^{M}(\text{pure})(M = \text{Au}, \text{Fe}). S_{\text{vib}}^{M}(\text{alloy})$ is estimated using the partial phonon density of states for respective elements at a given x. S_{vib}^{M} (pure) is the vibrational entropy of a pure element in its respective equilibrium phase. Configurational entropy of mixing can be expressed as $S_{\text{config}}(x) = -k_B[x \ln(x) + (1-x)\ln(1-x)]$. Figure 2 shows the concentration dependence of excess phonon entropy for $Au_{1-x}Fe_x$ at 300 K and configurational entropy of mixing. Square and triangle up (down) symbols indicate the total and partial vibrational entropies for Fe (Au). The inset shows the temperature dependence of excess vibrational entropy at various Fe concentrations. Clearly x = 0.19 is an anomalous point which separates the two unique regions of the phase diagram. In other words, the phonon entropy of mixing is negative for Fe concentrations $\leq 19\%$ beyond which it becomes positive. If we compare the configurational entropy of mixing, the calculated phonon entropy at x = 0.06 Fe is much larger and negative in sign. This implies that, up to

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Parameters	Pure Au	Au94Fe06	Au ₈₁ Fe ₁₉	Au ₇₅ Fe ₂₅	$Au_{50}Fe_{50}$	Pure Fe
C ₁₁ (GPa)	196.09 (201.63) ^a	192.22	222.01	205.04	211.29	328.35 (243.1) ^c
C_{12} (GPa)	164.14 (169.67) ^a	161.38	180.94	159.24	153.41	164.56 (138.1) ^c
C ₄₄ (GPa)	44.57 (45.44) ^a	48.69	57.64	61.53	81.75	136.00 (121.9) ^c
B (GPa)	174.79 (180.32)	171.66	194.63	174.51	172.70	219.16 (173.1)
G_V (GPa)	33.13 (33.65)	35.38	42.80	46.08	60.61	114.36 (94.1)
G_R (GPa)	25.97 (26.15)	26.13	33.46	36.74	47.26	107.57 (79.74)
$G_H = \mu$ (GPa)	29.55 (29.90)	30.76	38.13	41.41	53.94	110.96 (86.94)
Y (GPa)	83.93 (85.01)	87.07	107.37	115.12	146.56	284.82 (223.41)
C' (GPa)	15.98 (15.98)	15.42	20.53 (20.7) ^b	22.90	28.93	81.90 (52.50)
C_p (GPa)	119.56 (124.22)	112.69	123.30	97.71	71.66	28.56 (16.18)
ν	0.42 (0.42)	0.42	0.41	0.39	0.36	0.28 (0.28)
K_{ζ}	0.89 (0.89)	0.89	0.87	0.84	0.81	0.63 (0.68)
$A^{\dot{Z}}$	2.79 (2.84)	3.16	2.81	2.69	2.82	1.66 (2.32)
Р	5.92 (6.03)	5.58	5.10	4.21	3.20	1.98 (1.99)
λ (GPa)	155.09 (160.38)	151.15	169.21	146.90	136.74	145.18 (115.14)
$v_l (m/s)$	3335.20 (3377.67)	3372.61	3735.33	3675.10	4079.53	6523.11 (6058.51)
$v_t (m/s)$	1238.84 (1244.67)	1282.61	1472.16	1560.33	1915.67	3586.33 (3322.86)
Θ_D (K)	$162.87 (162.4 \pm 2)^{d}$	169.44	196.73	209.35	264.61	$539.29 (472.7 \pm 6)^d$

TABLE II. Calculated parameters for Au, Fe, and four alloys. Parenthetic values are measured data.

^aReference [33].

^bReference [34].

^cReference [35].

^dReference [36].

19% Fe, configurational entropy supports chemical mixing, but phonon entropy favors unmixing, predicting a miscibility gap in the alloy phase diagram [53]. Such discontinuity in the excess phonon entropy is attributed to the sudden uprise of $\Delta S_{\text{vib}}^{\text{Au}}$ at x = 0.25, which arises due to the stiffening of the Au-Au bonds in the vicinity of the Fe atoms. One also can explain this behavior from the enhancement of disorder broadening (see Fig. 1), a known fact for entropy enhancement. A similar abrupt change in excess entropy also is seen in the temperature dependence of ΔS_{vib} at x = 0.19(see the inset). Our calculated phonon entropy agrees fairly well with similar measured data published elsewhere [14].



FIG. 2. Excess phonon entropy vs x for $Au_{1-x}Fe_x$ at T = 300 K and configurational entropy of mixing S_{config} (blue curve). The inset shows ΔS_{vib} vs T at various x's.

Small discrepancies can be attributed to underestimation of the phonon DOS in experimental neutron weighted measurements.

In conclusion, we employ a different first-principles approach combining the SQS and ASR formalism to study the lattice dynamical and thermophysical properties of fcc $Au_{1-x}Fe_x$ alloys. This system is interesting because of the large differences in their constituent masses, force constants, and scattering lengths. In addition Fe, unlike in its elemental state, acquires a larger magnetic moment in the alloy. We found that, as the Fe concentration increases, the force constants tend to stiffen in the disordered environment. Above x = 0.19, phonon dispersion shows a split band behavior suggesting strong resonance that often arises due to dominant mass and/or force constant disorder. The anomaly at x = 0.19 is better described from our calculated phonon entropy, which suggests the possibility of chemical unmixing below 19% Fe and hence the onset of a miscibility gap in the phase diagram. Such an anomaly also is reflected in some of our calculated mechanical properties as well. As % Fe increases, size enhancement and force constant mismatches stiffen the material, which accounts for the increased Young's modulus and lower ductility. From the materials' perspective, the $Au_{1-x}Fe_x$ alloy is predicted to be mechanically stable and very ductile but highly anisotropic (the possibilities for microcracks are high). One of the main ideas of this Rapid Communication is to establish the combined SQS + ASR approach as an efficient and accurate method to study the lattice dynamical properties for random alloys.

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