

## Intermixing of a system with positive heat of mixing at high strain rates

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This paper investigates a mode of mechanically driven alloying of elements that are otherwise immiscible at temperatures well below the equilibrium melting point. Using molecular dynamics simulations, we have studied the alloying between Ag and Cu, a positive-heat-of-mixing ( $+\Delta H$ ) binary system with little solid solubility near ambient temperature, during high strain rate deformation at temperatures  $\leq 600$  K. Above a critical strain rate, both elements undergo mechanical melting into undercooled liquids, which are miscible at 600 K due to the reduced magnitude of the  $+\Delta H$  in the liquid state compared with that in the crystalline state. The nonequilibrium deformation maintains the melt state and assists the intermixing reaction between elemental Ag and Cu through stress-directed atomic flow. Upon unloading, the intermixed amorphous Ag-Cu crystallizes towards a supersaturated fcc solid solution. The scheme described is a process of low-temperature amorphization and intermixing of elements, and as such differs from the well-known liquid quench route that starts from an already-mixed liquid alloy at high temperatures.

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### I. INTRODUCTION AND STRATEGY

As seen in the equilibrium phase diagrams, a large number of binary metallic systems characterized by a positive heat of mixing ( $+\Delta H$ ) are virtually immiscible in the solid state near the ambient temperature.<sup>1</sup> This fact severely limits the opportunities to study the properties of the solid solutions formed by a given metal with various other elements. A well known nonequilibrium approach to create alloys (e.g., supersaturated solid solutions and amorphous phases) in such systems is to go to the melt or vapor phase and quench the alloy obtained at high temperatures to ambient temperature at very high cooling rates.<sup>2-4</sup> The formation of highly supersaturated solutions through the melt quench route has been demonstrated before for many  $+\Delta H$  systems including Ag-Cu.<sup>2,3</sup> Such an alloying mechanism may function even in some nominally “solid-state” processes such as ion irradiation,<sup>5</sup> shock compression,<sup>6</sup> or severe mechanical deformation,<sup>7</sup> where local regions can melt due to energy dissipation concentrated in thermal spikes or at “hot spots” under adiabatic conditions.

Here we demonstrate using molecular dynamics simulations a different mechanism for alloying a  $+\Delta H$  system, using Ag-Cu as an example. The strategy is to react two elemental melts, rather than two fcc crystals which would not react due to the absence of a thermodynamic driving force, at a temperature well below the equilibrium melting point. Normally an elemental liquid cannot be kept from crystallizing upon such large undercooling. A key ingredient in our strategy, therefore, is to create such melts at low temperatures mechanically through straining at very high rates. It is believed that Ag and Cu elemental liquids, if kept from crystallizing, can be mutually miscible down to fairly low temperatures. This is because the two elements are intrinsically much easier to mix in the liquid state than in the crystalline state. In the equilibrium solid state, fcc Ag and Cu crystals show a mutual solubility of no more than 1 at% below 600 K.<sup>1</sup> This low solid solubility originates from the positive  $\Delta H$  of the fcc solid solution due, in a large part, to the large size difference between Ag and Cu (13%). When

Ag and Cu are in the liquid or undercooled liquid state, atomic level stresses and strains due to atomic substitution are relaxed due to the changes in the local atomic arrangements and coordination number, such that the  $+\Delta H$  is much less positive. As a result of this reduced energy penalty to intermix liquid Ag and Cu, their liquid solubility is much higher than their solid solubility at all temperatures. In fact, at high temperatures with the help from the entropy of mixing liquid Ag and liquid Cu are well known to be completely miscible, as seen in the equilibrium phase diagram. An extrapolation of the existing enthalpy and entropy data suggests that liquid Ag and Cu can also be miscible over the entire composition range at a temperature as low as 600 K,<sup>8</sup> if all the solid fcc crystals can be prevented from forming and thus excluded from entering the picture. This is especially true if the liquid alloy develops short-range structures that further lower its free energy.

In our approach, melts of Ag and Cu will be produced through high rate straining; here heat dissipation is assumed to be so fast that there is *no* temperature increase during deformation, as opposed to the other extreme boundary condition of adiabatic heating. Such a constant temperature scheme is easily implemented in molecular dynamics (MD) simulations so that only dynamic loading effects are considered independent of the temperature effects.<sup>9</sup> The possibility of mechanically melting fcc crystals at ambient temperature by application of sufficiently large strain rates in uniaxial tension has been demonstrated very recently using MD simulations.<sup>9</sup> Other deformation modes are bypassed at such strain rates<sup>9,10</sup> so that above a critical strain the metal loses its shear rigidity due to the vanishing of shear modulus.<sup>10-12</sup> The instability triggers a homogeneous transformation to the amorphous state. For Ni, the critical strain rate is 5% ps<sup>-1</sup> and the resulting amorphous alloy has a calculated viscosity of 0.6 poise. In this work, we show that this strain-rate induced melting process also occurs in Ag and Cu upon uniaxial compression. We further demonstrate that the miscible Ag and Cu liquids are maintained under deformation, exhibiting deformation (stress)-assisted intermixing. Crystallization of the mixed amorphous Ag-Cu subsequent to defor-

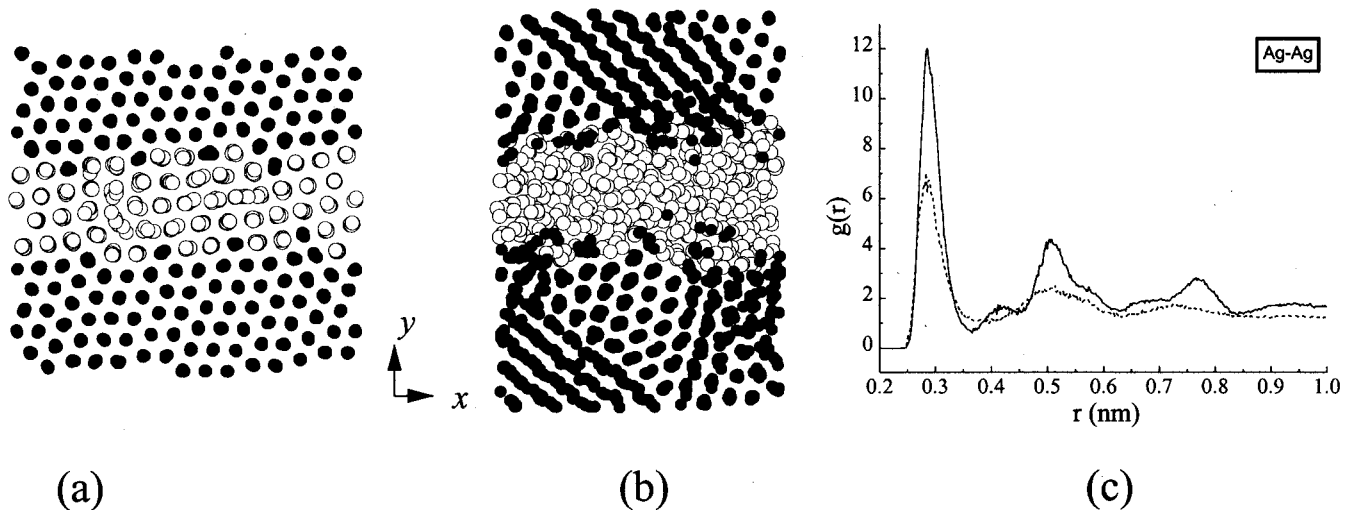


FIG. 1. (a) The initial Cu (full circles)/Ag/Cu multilayer configuration equilibrated at 600 K for 100 ps, (b) a snapshot of the same structure after 30% compressive strain along the  $x$  direction at a constant strain rate ( $1.0\% \text{ ps}^{-1}$ ), and (c) RDF of the amorphized Ag [dashed line, open circles in (b)] compared with that of fcc Ag (solid line).

mation leads to the formation of a supersaturated fcc solid solution. This is another example of externally driven alloying of immiscible elements under highly nonequilibrium conditions.

## II. METHOD OF SIMULATION

The MD model used consisted of a layer of fcc Ag (324 atoms) sandwiched between two layers of fcc Cu (1296 atoms). The thickness of the middle Ag layer, approximately 1 nm, was chosen to be larger than the cutoff distance of the potentials used in this simulation. Periodic boundary conditions were applied in all directions. Both the (100) planes and the [100] directions of the Cu and Ag crystals were set parallel. A (013) plane of Cu and a (031) plane of Ag were set to contact each other. Additional tests using other interface configurations showed little dependence of the mixing of Ag and Cu on the sample geometry (as explained below, amorphous structures develop first such that the initial crystal orientation does not have a strong influence on the alloying results). Simulations have been carried out in the constant pressure-constant temperature (Nosé-Parrinello-Rahman) ensemble. A fifth-order predictor-corrector algorithm with a time step of  $2 \times 10^{-15}$  s has been used to integrate the equations of motion.<sup>13</sup> We used the Nose-Hoover thermostat, where the system was coupled with an external heat bath by an extra degree of freedom. The internal temperature of the system during simulation was monitored and the temperature fluctuation was found to be no more than 50 K. (Note that, in experiments, the adiabatic heat dissipation during high-strain deformation would cause significant temperature increases.) Ag-Cu atoms interact via a many-body tight-binding potential developed recently by Mazzone *et al.*<sup>14</sup> The  $+\Delta H$  of the equiatomic liquid measured in an early experiment,  $+4.58$  kJ/mol at 1423 K relative to liquid Ag and Cu, was used as input in deriving this

potential. This value is reasonably close to the assessed value of  $+3.79$  kJ/mol.<sup>8</sup> We therefore decided to choose this potential since we are interested in the behavior of the liquid Ag-Cu alloys. Note that such a  $+\Delta H$  is much smaller than that for the fcc solid solution, calculated by various authors to be in the range of  $+6.8$ – $13.2$  kJ/mol relative to the fcc Ag and Cu references.<sup>7,8,14,15</sup> Using this same potential, the  $+\Delta H$  of the fcc solution is predicted to be as high as  $+13.2$  kJ/mol at the equiatomic composition.<sup>14</sup> As such, our model system is one that exhibits a large disparity in the magnitude of the  $+\Delta H$  between the liquid and the solid alloy. This is a feature that will highlight the strategy outlined in Sec. I.

## III. RESULTS AND DISCUSSION

Figure 1(a) displays the initial configuration of the fcc Cu/Ag/Cu sandwich equilibrated at 600 K. Intermixing between fcc Ag and Cu is minimal and remains so after long MD runs, in agreement with the known small equilibrium solid solubility. As shown in Fig. 1(b), deformation to 30% compressive strain along the  $x$  direction at a constant strain rate of  $1\% \text{ ps}^{-1}$  leads to predominantly cooperative shear events in Cu, while the Ag interlayer turns completely amorphous as confirmed in the radial distribution function (RDF) plot of Fig. 1(c). Cu also becomes entirely amorphous when the strain rate is increased to  $5\% \text{ ps}^{-1}$ . In comparison, Ikeda *et al.* observed a similar twinning mode at or below a strain rate of  $0.5\% \text{ ps}^{-1}$  in Ni, which amorphized at  $5\% \text{ ps}^{-1}$ .<sup>9</sup> The critical strain rate for amorphization is the lowest for Ag probably due to its comparatively low melting point.

In Fig. 1(b), some intermixing of Ag and Cu is already noticeable near the interfaces where Cu begins to amorphize. The intermixing becomes more obvious at higher strain rates when both metals melt (see below), increasing with total strain (time for interdiffusion). At large strains and strain rates, however, the shape of the MD box changes signifi-

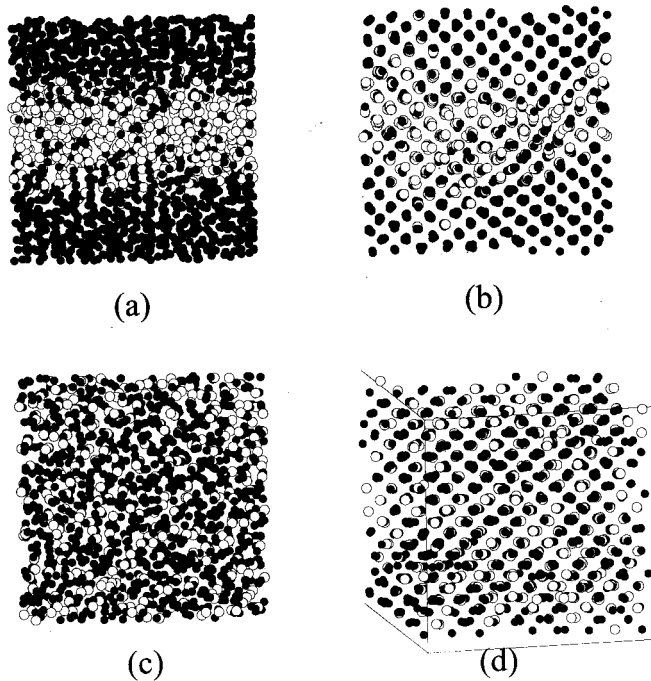


FIG. 2. Snapshots of the Cu (full circles)/Ag (open circles) system, showing (a) the full amorphization of Ag and Cu and intermixing after two cycles of deformation, (b) crystallization after subsequent relaxation for 100 ps, (c) extensive and uniform mixing after two additional cycles, and (d) the final random solid solution structure after relaxation.

cantly, with dramatic elongation along the  $y$  direction and shrinkage in the  $x$  direction (Fig. 1). To prolong straining to observe more mixing, we performed cyclic loading in both  $x$  and  $y$  directions (1 cycle=1 loading along  $x$  and 1 along  $y$ ). Alternative to constant strain rate, we used loading at a fixed increment of 0.5 MPa per time step in the  $x$  direction. The stress-strain curve is monitored during loading, and so is the strain rate. After the strain rate reaches  $5\% \text{ ps}^{-1}$ , at which point the total strain is in the 30–40% range, the external stress is withdrawn at 5 MPa per time step. The same operation is then performed along  $y$ . The simulation box regains a shape similar to that at the outset of the straining. Figure 2(a) shows the system configuration at the end of the second cycle. It is readily seen that due to the high strain rates reached, now both Cu and Ag are melted, and the penetration of them into each other is much more pronounced than in Fig. 1(b). There appears to be more mixing of Cu into Ag than vice versa. This is probably because Ag has a lower melting point so it melts earlier than Cu and is relatively less viscous, such that there is more Cu incorporation through liquid diffusion. Signs of these can be found in Fig. 1. Also, we noticed that in the equilibrium phase diagram, at low temperatures Cu has a larger equilibrium solubility in Ag than vice versa. Similar asymmetric scenario may exist in the nonequilibrium undercooled liquid case. After the dynamic loading is over, the system solidifies and crystallizes. Figure 2(b) shows the configuration of the system in Fig. 2(a) after subsequent relaxation for 100 ps. The unreacted elements

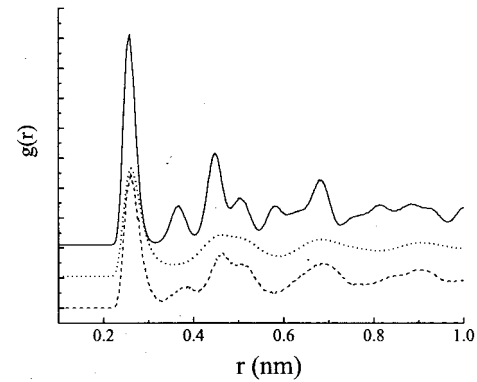


FIG. 3. RDFs of the alloys in Fig. 2(c) (dotted line) and Fig. 2(d) (dashed line), compared with that of pure Cu (solid line), showing the intermixing and amorphization as well as the subsequent crystallization into an fcc supersaturated solid solution after loading.

have now returned to elemental fcc crystals. The intermixed amorphous layer, on the other hand, crystallizes into a non-equilibrium alloy. This alloy is an fcc supersaturated solid solution, to be discussed below.

Even more intermixing is observed after four deformation cycles as shown in Fig. 2(c), due to more straining introduced during the cycling and more time for interdiffusion. It is obvious that now Cu atoms have spread throughout the Ag layer, and vice versa. The intermixing is clearly much more pronounced and uniform than that in Fig. 2(a) and 2(b). The intermixed layer is now a concentrated liquid (amorphous) alloy, as demonstrated by the features of the broad halos in the RDF plot shown in Fig. 3 (compare the halos with those peaks of pure metals). Upon unloading, the amorphous interlayer again begins to crystallize towards the fcc structure, as is evident in the RDF shown in Fig. 3. The alloy is a supersaturated fcc solid solution, as suggested by the peak shifts in the RDF, i.e., the changes of the interatomic distances relative to pure Ag or Cu. In fact, the uniform and random mixing of Ag and Cu in a twelve-coordinated fcc structure is confirmed through an analysis of the nearest neighbor bonds for the snapshot in Fig. 2(d), by counting the Cu-Cu, Cu-Ag, and Ag-Ag bonds corresponding to the first RDF peak. Such a bond analysis for Cu shows that, on average, the number of like bonds is 9.4 and that of unlike bonds is 2.6. The percentage of Ag neighbors (21.6%) corresponds very well to the statistical average expected for a random solution alloy of our sample configuration (23% Ag). Similarly, on average each Ag has 9.4 unlike neighbors and 2.6 like neighbors. Potential energy calculations using MD confirmed that the crystallized fcc solution state had a lower energy than the amorphous structure created by deformation.<sup>14</sup> This is consistent with the experimentally observed transformation of amorphous Ag-Cu, once formed by coevaporation onto cold substrates, into a single-phase fcc solution at a fairly low annealing temperature (370 K) prior to decomposition into Ag and Cu.<sup>16</sup> However, due to the kinetics required, in simulations it is difficult to fully complete the crystallization into fcc solution on MD time scales, more so in the case in Fig. 2(d) than the less concentrated alloys in Figs. 2(a) and 2(b).



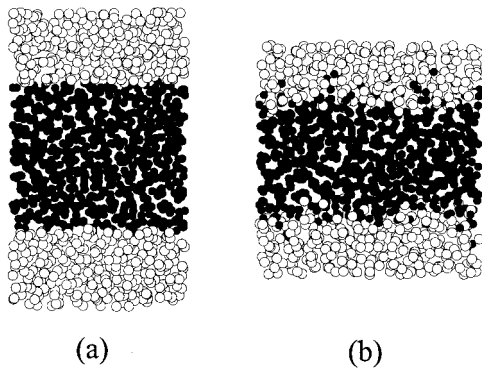


FIG. 4. Quenched liquid Cu (full circles) and liquid Ag (open circles) in contact, (a) as-constructed and (b) after 30% of compressive strain along the  $y$  direction at  $1\% \text{ ps}^{-1}$  at 600 K.

Therefore, residuals of amorphous structure and unsettled atomic positions seem to remain at 600 K in Fig. 2(d), and the resulting RDF is not yet that of a perfect fcc single phase solution, Fig. 3. Our additional MD result also confirmed previous MD simulation findings that amorphous Ag-Cu can be obtained at room temperature in the central concentration range<sup>17</sup> by using extremely rapid MD quench of a liquid Ag-Cu alloy. However, we see here that the fcc solution is the lower enthalpy phase. This more stable phase forms instead of the competing amorphous alloy if a lower MD quench rate is used, or if the liquid alloy is quenched to and held at an intermediate temperature such as 600 K.

In addition to creating the melts by deformation of crystals, one can also obtain and retain the elemental Ag and Cu liquids by rapid quenching from 2000 to 600 K in the computer.<sup>17</sup> To observe and confirm that undercooled liquid Ag and Cu would react, these undercooled elemental melts have to be kept from crystallizing. This again can be done by deformation. In this simulation, the Ag/Cu/Ag sandwich is formed by bringing together undercooled Ag and Cu, with an interspacing chosen to minimize cohesive energy at zero temperature. As expected, without straining, elemental liquids cannot be sustained when being held at this temperature. However, as shown in Fig. 4, after 30% compressive strain along  $y$  at a fixed strain rate of  $1\% \text{ ps}^{-1}$ , both elements remain amorphous and the intermixing between Ag and Cu is observable. The amount of mixing is much less than that in Fig. 2; this is because in this case only one action of straining was performed, rather than the multiple cycles used in Fig. 2. It appears that in this case  $1\% \text{ ps}^{-1}$  is sufficient to keep the majority of the system from crystallizing for the duration of this computer experiment. As a comparison, without straining the Ag and Cu layers crystallize quickly, becoming fully crystalline within 100 ps. Only slight intermixing was observed to occur between Ag and Cu before the completion of crystallization. Note that here again more Cu is entering Ag than vice versa, confirming that this asymmetry seen in Fig. 2 is not due to the relatively small thickness of the Ag layer used there.

In addition, the internal stress is monitored as a function of strain. After the stress reaches a plateau,<sup>9</sup> the viscosity of the assembly is estimated (stress divided by strain rate) to be

on the order of 1 poise. This value is similar to that of the amorphous Ni at  $5\% \text{ ps}^{-1}$ .<sup>9</sup> Such viscosity values for our undercooled liquids are to be compared with those at the equilibrium melting points of these metals, about 0.04 poise.<sup>18a</sup>

Several observations can be made from the results presented above. First of all, liquid Ag and Cu are apparently miscible at 600 K, while no observable intermixing between fcc Ag and fcc Cu is expected at this temperature. The amount of diffusion seen can be compared with the liquid diffusivity extrapolated from the known Arrhenius laws<sup>18b</sup> or estimated based on its inverse dependence on viscosity (Stokes-Einstein relation).<sup>19</sup> Such miscibility and interdiffusion are expected from the increased free volume and reduced magnitude of the  $+\Delta H$  in liquids. Secondly, high rate straining directly drives fcc Ag and Cu into the undercooled liquid state at a constant temperature way below the equilibrium melting point. While elemental liquid Ag and Cu crystallize quickly at 600 K, high strain rate deformation maintains the miscible melts, greatly facilitating the intermixing. Thirdly, under high-rate flow, the melts exhibit a viscosity expected for liquids at such a temperature. Moreover, the nonequilibrium external forcing introduces shearing forces and stress (strain) gradients in the undercooled melts, causing stress-directed atomic flow favoring intermixing.<sup>20,21</sup> MD simulations have suggested that the local heterogeneous stress field at the interface in the disordered media can make each atom feel a different value of the net force.<sup>22</sup> Such a force can drive each individual atom, one at a time, to jump pass its saddle point configuration separating two possible locations.<sup>22</sup> Each of this localized, random process contributes a minute amount to the overall strain as well as intermixing, superimposing on and modifying the normal thermally activated diffusion. As such, the enhanced mixing observed is believed to be a result of deformation (stress)-assisted interdiffusion. Finally, after the deformation is over, the intermixed Ag-Cu melt freezes and crystallizes into a supersaturated fcc solid solution in a way similar to quenching from the high-temperature liquid in laboratory experiments.<sup>2,3</sup> This fcc alloy results because it has a lower energy than the amorphous alloy,<sup>14</sup> and is relatively easy to attain because it comes from the amorphous alloy almost in a partitionless fashion. Phase separation of the solid solution, driven by its positive heat of mixing, into the ground state pure fcc Ag and Cu would require partitioning in the solid state and thus prolonged annealing at 600 K way beyond what is realizable in MD simulations. For concentrated alloys, even the amorphous state may be (partially) retained at room temperature after a rapid MD quench.<sup>17</sup>

Enhancement of solubility well above equilibrium values can also be observed at lower temperatures, e.g., at 300 K, but with smaller solubility and intermixing rates. This observation suggests that the intermixing observed is due to both thermally (subject to chemical driving force) and mechanically activated atomic movements. The thermodynamic driving force for interdiffusion in the liquid diminishes with decreasing temperature as the positive, albeit small, heat of mixing becomes increasingly dominant. At sufficiently low temperatures, the effects of the entropy of mixing are over-

whelmed by the positive heat of mixing and intermixing has to come entirely from mechanically driven mixing against the demixing flux driven by the now unfavorable thermodynamic driving force. Note again that our alloying scheme is different from the conventional liquid quench route which starts from thermodynamically melted and already-mixed equilibrium liquid alloy at high temperatures above the melting point.

The extremely high strain rates used in this study, although at the high end of and even beyond those observed in shock wave and high velocity impact experiments, are typical for MD simulations.<sup>9,17</sup> However, caution must be exercised if one is to attempt to compare the MD findings with alloying in real-world experiments such as milling or cold rolling. For example, in mechanical alloying experiments using ball milling, the average strain rate is much lower (e.g.,  $10^4/s$ ) so that other plastic deformation mechanisms in crystals, such as dislocation activities, twinning, and void formation, may preempt strain-induced (local) melting or amorphization. Also, in real experiments adiabatic heating is difficult to avoid at high strain rates inside shear bands. On the other hand, due to the heterogeneous nature of the plastic deformation and the deformed microstructure, there may be local regions where amorphouslike structures are present, such as at the cold welded Ag/Cu interfaces or high-energy grain boundaries created during severe plastic deformation (milling or rolling). In fact, local amorphous regions have been observed in heavily cold worked Ag-Cu,<sup>23</sup> and amorphous alloys can form even in immiscible systems.<sup>24</sup> In such cases, deformation may proceed in a way similar to what is observed above at the interfaces, or inside amorphous regions, and cause relative motion of contacting grains<sup>22</sup> or intermixing of neighboring atoms.

#### IV. SUMMARY REMARKS

We note that the two interacting elements in our MD model and potentials do not have to be taken literally as silver and copper. In essence, we are modeling a  $+\Delta H$  system that has a fairly large atomic size mismatch and hence a much larger  $+\Delta H$  in the solid than in the liquid.

In summary, our MD simulations allowed the decoupling of dynamic structural/chemical changes from mechanisms associated with temperature spikes and heat transfer. At ultrahigh strain rates, conventional deformation modes are bypassed and strain-induced amorphization/melting of metals provides an avenue for alloying of elements otherwise immiscible at low temperatures. During such deformation, metal melts are created and maintained. Different from the crystalline state where there is a relatively large strain energy penalty associated with alloying size-mismatched Ag and Cu, in the more accommodating liquid state elemental Ag and Cu are easier to mix and hence miscible at a temperature as low as 600 K. Under these mechanically driven conditions, which also introduce localized, stress-directed, random atomic motions in a disordered media,<sup>22</sup> significant intermixing occurs at 600 K within very short time, forming Ag-Cu alloys far beyond the equilibrium solubility. While the amorphous state of the metals and the intermixed layer is only transient at 600 K, after dynamic loading a nonequilibrium binary alloy is obtained through crystallization towards a supersaturated fcc solid solution. Both the amorphous and the solid solution phases can be retained if a rapid quench to room temperature is implemented.<sup>17</sup>

Taken together with our earlier work on Ag-Cu intermixing on free surfaces,<sup>15</sup> our studies show examples of new schemes designed to enhance solid solubility in  $+\Delta H$  systems at low temperatures. Both of our schemes take advantage of the structural configurations that lower the magnitude of the positive  $\Delta H$ . At the same time, these configurations also open new kinetic pathways that allow fast mixing at fairly low temperatures; the mixing is realized either through atoms swapping positions in relatively open regions near free surfaces, or through stress-biased/assisted interdiffusion to result in random mixing in the viscous flow of an undercooled melt under dynamic and externally driven conditions.

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