Twinnability of bimetal interfaces in nanostructured composites

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Bimetal interfaces hold the extraordinary potential to promote or suppress deformation twinning in nanostructured composites. This article constructs a methodology for developing maps for identifying the twinnability of chemically sharp, bimetal interfaces based on their structure and properties. The map is shown capable of rationalizing the variation in experimental observations among several different bimetal interface structures.

\textbf{Keywords:} Interfaces, Twinning, Nanomaterial, Composites, Nucleation

Deformation twinning is one of the few basic mechanisms that face-centered cubic (fcc) metals \cite{1–3} employ to plastically accommodate an applied mechanical force. For single-phase fcc metals, there has been widespread interest in understanding the nucleation and growth of deformation twins and establishing guidelines for determining which materials can twin and when and where in the material twins form \cite{4,5}. The appearance of twins can be correlated to applied conditions, such as low temperatures and/or high strain rates, in which the thermodynamic processes that underlie slip dislocation motion become difficult \cite{6,7}, or to material areas experiencing high localized stress concentration, such as near surfaces or cracks \cite{8,9}. Also playing a role are chemical and physical material properties, such as the intrinsic stacking fault energy, other maxima and minima on generalized planar fault energy (GPFE) curves, alloying, and crystallographic orientation \cite{1,6,10–13}. Within suitably oriented fcc crystals, twinning occurs more readily in metals with a low stacking fault energy (SFE), such as Ag (intrinsic SFE $\gamma = 16 \text{mJ/m}^2$ \cite{14}) than those with an intermediate SFE, such as Cu ($\gamma = 45 \text{mJ/m}^2$ \cite{15}) or a high SFE, like Al ($\gamma = 122 \text{mJ/m}^2$ \cite{2}). In addition to the above, as grain sizes reduce to the nanoscale, grain size becomes an important factor in the promotion of deformation twins, and twins originate from grain boundaries \cite{4,10,16–19}. Within the nanoscale range, an optimal grain size has been observed, below which twinning becomes more difficult with decreasing grain size \cite{20,21}. However, when deformation twins originate from bimetal interfaces, observations vary widely, from completely suppressed twinning \cite{22} to fine twins with numerous twin boundaries (TBs) \cite{23,24} to fully twinned structures with no visible TBs \cite{25,26}. The prior guidelines outlined above cannot rationalize the complete lack of twinning in 30 nm Cu within rolled nanolayered physical vapor deposition (PVD) Cu–Nb composites \cite{22} or profuse twinning in submicron Cu within Cu–Ag composites \cite{25} or profuse twinning in submicron Cu within Cu–Ag composites rolled at elevated temperatures \cite{25}. These contradictions point to a need to revise twinning guidelines when considering two-phase materials. In this letter, we present an approach for developing a twinnability map for twinning from bimetal interfaces based on the interface structure. The interfaces considered here are sharp, exhibiting no chemical variation. All the above-mentioned conditions for deformation twinning still apply (suitable GPFE, crystal orientation relative to an applied stress, grain size and high stresses and/or rates), but are not sufficient without account of the structure of the interface.

When two crystals, differing in lattice parameters and/or in crystal structure, are joined, the relaxed equilibrium structure of the common interface that forms has

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an atomic structure that is unlike that of either crystal [27–30] (Figure S1(e)–(h)). The resulting interface can adopt a coherent structure (containing no intrinsic interface dislocations) or a semi-coherent structure (containing discrete interface dislocations) or an incoherent structure (often a disordered structure, in which discrete interface dislocations cannot be identified). Because coherent and incoherent interfaces do not have preferred nucleation sites for lattice dislocations, we focus on semi-coherent interfaces in the present work. Semi-coherent interfaces possess a well-defined, ordered atomic pattern, described as a regular network of discrete interface dislocations [30–32] (Figure S1). The network contains $n$ arrays of discrete interfacial dislocations $(i)$, $i = 1, \ldots, n$ (see Figure 2(a), where $n = 2$). The number $n$ of distinct arrays, their Burgers vectors $B^{(i)}$ and spacing $S^{(i)}$ for a given interface can be calculated using a combination of atomic-scale simulation (Figure S1(d)–(e)) and analytical methods described in [31–33] (Table 1 in supplement). 

Within this class of semi-coherent interfaces lies a rich set of structures, not all of which can twin. The strong influence of interface structure is evident in Figure 1(a), which displays a transmission electron microscopy (TEM) micrograph of a shocked Cu–Nb sample [34], wherein the semi-coherent interface in the center forms twins (Figure 1(c)), but those on the left did not. The corresponding high-resolution TEM (HRTEM) micrographs of this area shows that the twinned interface was \{112\}||\{111\}Cu\{110\}Nb, which is a faceted interface (Figure 1(d)), while the untwinned interface was \{111\}||\{110\}Cu\{110\}||\{111\}Nb, an atomically flat one (Figure 1(b)). Observations of twins from the former interface as well as other semi-coherent faceted interfaces are shown in Figure S1(a)–(c).

For the twinnability of these semi-incoherent interfaces, two mechanisms for successive emission of twinning dislocations (TDs) are considered. TDs can be emitted from an intrinsic interface dislocation or from an extrinsic interface dislocation. The former ‘intrinsic’ mechanism can involve, for instance, a non-planar dissociation of an intrinsic dislocation [35,36]. Should this be the dominant mechanism, the faceted interface (Figure 1(d) and S1) would be favored over the atomically flat one. The former interface has interface dislocations with in-plane and out-of-plane Burgers vectors, i.e. $B^{(i)} = B^{(i)}_p + B^{(i)}_n$, which could supply both Burgers vector components needed for a TD, unlike the flat interface, which only contains interfacial dislocations with in-plane Burgers vectors, i.e. $B^{(i)} = B^{(i)}_p$ (Figure 1(b)). The latter ‘extrinsic’ mechanism can be identified as an interface slip–twin transmission event, wherein a glide dislocation (an extrinsic dislocation) runs into the interface from the adjoining crystal, and finally emits into the fcc crystal as a TD [29,37,38]. The mapping methodology proposed below aims to predict the potential of a bimetal interface structure to act as a source of TDs via these two basic mechanisms for TD emission.

The first is referred to here as the dissociation (or D) mechanism. Under a mechanical driving force, TDs can emit directly from the interface via either dissociation of an interfacial dislocation [30] or indirectly from local stress concentrations generated around the interfacial dislocations [39]. The D mechanism that we consider hereinafter corresponds to the former, since its activation barrier is much lower than that for the latter and hence is the more likely of the two to initiate the twinning process.

Twin formation via the D mechanism is proposed to proceed in two steps. In the first step Figure 2(b), a TD is produced via dissociation of an interfacial dislocation, prompted by stress. Dissociation reactions should obey a few constraints, e.g. see [36], one of which translates here to a repulsive interaction between the residual dislocation left at the interface and the TD. In addition, energetically favored dissociation reactions are also those that reduce
nucleation of a TD on every twinning plane within $s^{(i)}$. If $k$ planes lay in-between then $k$ TDs need to nucleate (Figure 2(c)). When $k$ equals one, then nucleation is spontaneous and a three-layer twin will form [17], but when $k$ is too large, then twinning via the D mechanism is not possible. (The stacking faults SF1 and SF2 behave instead as individual partials, from which twinning by other mechanisms have been proposed [4,20].) Whether or not the needed $k$ TDs can nucleate can be determined via the following energetic expression for the change in energy due to the nucleation of $k$ TDs:

$$
\Delta E = kE_{\text{self}} + \frac{(k + 1)d}{\sin \theta} \Delta E_{\text{int}} - E^\text{int} - W_{\text{strain}}(K)
$$

where $d$ is the inter-planar spacing for the twin plane, $\theta$ is the angle between twin and interface plane, and $l_k$ is the distance traveled by the $k$ TDs (Figure 2(c)). The first term is the self (core) energy of the $k$ TDs, the second $\Delta E_{\text{int}}$ is the change in the interface energy per length of twinned interface, the third is the reduction in energy due to the removal of the two interior stacking faults, the fourth is the interaction energy between the oppositely signed $k$ TDs created upon nucleation, and the last term is the strain energy supplied by the glide $k$ TDs. Applying Equation (2), a critical $k^*$ can be defined such that nucleating more than $k^*$ TDs is unfavorable. We estimate in the supplement that for nanolayered Cu ($k < 100$ nm), twin formation under the D mechanism is possible for interfacial structures in which $k^* \simeq 2$ (Figure S2). This value $k^*$ corresponds to a critical spacing $s^*$ between two adjacent dissociation events, $S^* = d(k^* + 1) / \sin \theta$. According to this analysis, the likelihood that array (i) can support both steps in the sequence can be mathematically expressed by a product of the ratio in Equation (1) and ratio of the critical spacing to the actual one $s^{(i)}$

$$
S^{(i)} = \left( \frac{B_{N}^{(i)}}{b} \right) \left( \frac{(d(k^* + 1))}{(s^{(i)} \sin \theta)} \right)
$$

Satisfying criteria for both steps to proceed translates to $S^{(i)} \geq 1$. If either $s^{(i)}$ is too large or the value of $B_{N}^{(i)}$ is too small, then twinning via the D-mechanism from array (i) is unlikely. For an interface containing more than one array of interfacial dislocations, its twinning propensity can be represented by the array (i) with the largest $S^{(i)}$, denoted as $S_{\max}$

$$
S_{\max} = \max_i S^{(i)}
$$

A TD can also be supplied to the fcc crystal by slip–twin interface transmission, wherein a lattice dislocation from the adjoining crystal impinges on their common interface, becomes absorbed into the interface, and emits into the fcc crystal as a TD. We refer to this second mechanism.
as the $T$ mechanism. Slip–twin transmission is affected by both the core configuration of the lattice dislocation in the interface and the geometry of crystallographic slip across the interface. When absorbed in the interface, the lattice dislocation changes its core structure in response to the atomic structure of the interface. The degree of in-plane core spreading is inversely proportional to the interfacial shear strength ($\tau_{\text{ISS}}$) [40], the threshold in-plane shear stress at which the two crystals begin to slide relative to one another. A relatively low $\tau_{\text{ISS}}$ encourages the lattice dislocation to spread within the interface plane, which hinders the slip-transmission process. Independent of core spreading, slip–twin transmission is also affected by the extent to which the active incoming slip system $\alpha$ and outgoing $\beta$ twin system are aligned. Experimental studies on grain boundaries find that the relative orientation of these two systems can be effectively quantified by $F \equiv \left(\cos \left(\frac{\pi}{2} \frac{\Psi_{\alpha\beta}}{\psi_c}\right)\right) \left(\cos \left(\frac{\pi}{2} \frac{\theta_{\alpha\beta}}{\theta_c}\right)\right) \left(\cos \left(\frac{\pi}{2} \frac{\theta_{\alpha\beta}}{\theta_c}\right)\right) [41], where $\Psi_{\alpha\beta}$ is the angle between the Burgers vectors and $\theta_{\alpha\beta}$ is the angle between the glide plane and interface intersection lines made by $\alpha$ and $\beta$. The brackets $\langle \rangle$ signify zero when either $\Psi_{\alpha\beta}$ or $\theta_{\alpha\beta}$ exceeds a limit $\psi_c$ or $\theta_c$, and $x$ otherwise. To account for the applied driving force, we multiply $F$ by the Schmid factors $\left[m_\alpha \right]$ and $\left[m_\beta \right]$ of slip $\alpha$ and twin $\beta$, with the additional conditions that twin $\beta$ must satisfy twinning directionality and incoming and outgoing $\alpha$ and $\beta$ systems share the same line orientation $\xi$, as indicated by the $\left[ \xi \right]$ brackets (see supplement). On this basis, assessing the effectiveness of a given slip–twin transmission pathway for twinning is given by

$$G = \left(\cos \left(\frac{\pi}{2} \frac{\Psi_{\alpha\beta}}{\psi_c}\right)\right) \left(\cos \left(\frac{\pi}{2} \frac{\theta_{\alpha\beta}}{\theta_c}\right)\right) \left[\cos \left(\frac{\pi}{2} \frac{\theta_{\alpha\beta}}{\theta_c}\right)\right] \left[m_\alpha \right] \left[m_\beta \right], \quad (5)$$

The maximum value of $G$ in Equation (5) is $G_0 = 0.25$. The most effective pathway offered by the interface is associated with the highest value of $G$ among all possible $\alpha$–$\beta$ pairs, denoted as $G_{\text{max}}$. In the calculation of $G$ and $G_{\text{max}}$, we used $\theta_c = 15^\circ$ and $\psi_c = 45^\circ$ [41]. Geometrically efficient pathways can be expected to have a value of $G_{\text{max}} \geq 0.0156$ (or $G_{\text{max}} / G_0 \geq 0.0625$), which corresponds to good geometric alignment $\left(\cos \left(\frac{\pi}{2} \frac{\Psi_{\alpha\beta}}{\psi_c}\right)\right) \left(\cos \left(\frac{\pi}{2} \frac{\theta_{\alpha\beta}}{\theta_c}\right)\right) \left[\cos \left(\frac{\pi}{2} \frac{\theta_{\alpha\beta}}{\theta_c}\right)\right] \geq 0.25$ and large Schmid factors, each 0.25. Interfaces in which all favored pathways connect to the same twin plane ($N = 1$), such as the $\{112\}(111)f_{\text{cc}}||\{112\}(110)\text{bcc}$ interface (Figure S3(d)), present an instability that favors twin nucleation and thickening. Some interfaces may present more than one favorable dislocation slip transmission pathway ($2 \leq N \leq 4$). Unlike the case of $N = 1$, multiple pathways ($2 \leq N \leq 4$) can potentially stabilize the adjoining crystal and hinder instabilities such as twinning (Figure S3(a)–(c)). We account for this effect later.

For demonstration, we apply the above measures to Cu–Nb interfaces deformed in plane strain compression normal to the interfaces (unless stated otherwise). Cu is the fcc metal in which twin formation is questioned and Nb is the bcc metal to which it is joined. To remove the influence of grain size in this example, we consider samples in which the individual Cu layers are one-grain thick and nanometer in scale. We begin with semi-coherent interfaces found in two Cu–Nb experimental systems that vary significantly in their intrinsic atomic structure as a result of differences in fabrication history. Set 1: Cu–Nb multilayers with nanolayer thicknesses ($h < 100$ nm) fabricated by accumulative roll bonding (ARB), which are dominated by $\{112\}(110)\text{Cu}||\{112\}(111)\text{Nb}$ interfaces within $10^\circ$ [42] (Figures 1(d) and S1(a) and (c)). Set 2: Cu–Nb multilayers with nanolayer thicknesses fabricated by PVD, which develop thermodynamically equal portions of Kurdjumov–Sachs (KS) and Nishiyama–Wasserman (NW) interfaces [42,43] (Figure 1(b)). These have also been found to be mechanically stable with strain, provided starting layer thicknesses are less than $100$ nm [44].

The tendency for transmission can be determined by mapping interfaces on a 2D plot of $\tau_{\text{ISS}} / \tau_{\text{theo}}$ vs. $G_{\text{max}} / G_0$, such as in Figure 3. The $\tau_{\text{ISS}}$ is normalized by its upper limit, the theoretical shear strength $\tau_{\text{theo}}$, and reporting a single value suffices.
In this map, interfaces that would lead to twinning via the T mechanism must exhibit both high $r_{ISS}/r_{theo}$ (no core spreading) and high $G_{max}/G_0$ (geometrically effective pathway(s)), $\geq 0.0625$ and hence lie in the upper right-hand region. We apply the map in Figure 3 to a few experimental observations of twinned (black) and non-twinned (blue) interfaces, using $r_{theo} = 2.6$ GPa for Cu and values for $r_{ISS}$ from atomic-scale simulation (Table 1 in the supplement). Considering twin formation via the T-mechanism alone, twinning can be associated with interfaces with a large value of $\chi = (r_{ISS}/r_{theo}) (G_{max}/G_0)/N$, where $N$ is the number of effective transmission pathways. Having $N = 1$ such as in Figure S3(d) promotes a dynamic instability that favors twinning, whereas multiple pathways ($N > 1$) such as in Figure S3(a)–(c) would not.

The propensity as well as the mechanism for twinning can be determined with the TD source map on Figure 4, where one coordinate is related to the D mechanism and the other to the T mechanism $\chi$ from Figure 3. The boundary $S_{max} = 1$ marked in Figure 4 corresponds to the limit below which twin formation via the D mechanism alone is not possible. The map is designed such that twinning is unlikely for interfaces that lie close to the origin of this map, where the possibilities of operating the D and T mechanisms are low. As confirmation, interfaces in which twinning was not observed (blue points) collect in the ‘no-twinning’ region of the map. Two of these are the KS and NW interfaces, which are found in PVD nanolayered composites. Studies show that no twins are observed from these interfaces, regardless of layer thickness (even down to 5 nm), strain level, and strain rate (from quasi-static rolling to gas gun to laser shock [22,34]). In all these tests, the samples were compressed normal to the interfaces. The map indicates that if these interfaces were loaded instead in normal tension (diamonds), they still would not twin.

The propensity for twinning becomes more favorable with distance away from the origin. Twinning is especially likely for interfaces falling in the upper right-hand region of the map, where both the D and T mechanisms are favored. One interface that belongs to this twinning class is the $\{112\}⟨111⟩$Cu||$\{112\}⟨110⟩$Nb interface, which has been reported in many studies to produce twins on a particular twin plane [23,34] (Figure S1(a)). Significantly, twins in layers as large as 200 nm are observed to form from these interfaces [26]. The map indicates that this interface uses both D and T mechanisms to nucleate and expand twins on the same twin plane. Another interface found in the Set 1 composites was the faceted $\{338\}⟨443⟩$Cu||$\{112\}⟨110⟩$Nb interface, corresponding to an energy cusp in the formation energy landscape [32] (Figure S1(f)). Substantially less twins have been associated with this interface (Figure S1(c)). Its placement on the map suggests that it twins predominantly by the T-mechanism.

We utilize this map to predict the twinnability of other interfaces. We begin with interfaces occurring less frequently in the Set 1 material, such as the faceted $\{112\}⟨111⟩$Cu||$\{001\}⟨110⟩$Nb interface (Figure S1(e)). The map predicts that this interface can twin using both D and T mechanisms. In agreement, we observe that twins are just as likely to form from $\{112\}Cu||\{001\}$Nb interfaces (Figure S1(b)). Next we consider another set of ARB material, Set 3, with extremely fine nanoscale thicknesses ($h \leq 20$ nm), and different predominant interfaces, e.g. $\{551\}⟨1\ 1\ 10⟩$Cu||$\{112\}⟨111⟩$Nb interface and the $\{110\}⟨001⟩$Cu||$\{112\}⟨111⟩$Nb interface [26]. Twins are not often observed from these interfaces. According to the map in Figure 4, they are not expected to twin. As a last example, we examine a fourth set of Cu–Nb nanolayers ($h \leq 30$ nm) formed by rolling the Set 1 samples transversely, forming mainly an atomically flat $\{110\}⟨111⟩$Cu||$\{001\}⟨110⟩$Nb interface. Twin formation was not associated with this interface (Figure S1(d)). This interface lies in the no-twinning region of the map since it is atomically flat (no D mechanism) and it favors dislocation transmission on multiple pathways (high $N$), which favors crystal stability and hinders instabilities, such as twinning (or shear banding).

The primary role of the twinnability map proposed here is to determine whether or not a given interface structure under a prescribed stress state can successively
emit TDs. For sufficiently fine nanostructures, this critical nucleation step can be correlated with a measure _twinning propensity_. The map reveals the structural features controlling twinning and the type of twinning mechanism they drive. To date, it has been the only successful way to rationalize seemingly inconclusive observations on nanolayered composites. The map adds, and does not replace, an essential factor to previously established guidelines for twinning in fcc metals. To isolate the effects of interface structure from layer size $h$, strain rate and temperature, we fixed the latter set of variables. All experimental observations used for validation had nanoscale layer thicknesses ($100 \text{ nm} < h < 10 \text{ nm}$). Like the leading partial, interface structure is also suspected to govern nucleation of the trailing partial, possibly giving rise to interface-structure-dependent grain-size effects. Size effects will be considered in future extensions of the map. The framework introduced here is suitably fundamental and can be applied to other bimetal interfaces involving different crystal structures, e.g. fcc/fcc and fcc/hcp. Bimetal interface twinnability maps can open the way to tuning bimetal interfaces to govern the relative activities of slip and twinning, the key to controlling microstructural evolution and material response under deformation.

**Supplementary online material.** More detailed information on the theory and experiments are available at http://dx.doi.org/10.1080/21663831.2013.782074.

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