



# Influence of slip and twinning on the crystallographic stability of bimetal interfaces in nanocomposites under deformation

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## Abstract

In this work, we examine the microstructural development of a bimetal multilayered composite over a broad range of individual layer thicknesses  $h$  from microns to nanometers during deformation. We observe two microstructural transitions, one at the submicron scale and another at the nanoscale. Remarkably, each transition is associated with the development of a preferred interface character. We show that the characteristics of these prevailing interfaces are strongly influenced by whether the adjoining crystals are deforming by slip only or by slip and twinning. We present a generalized theory that suggests that, in spite of their different origins, the crystallographic stability of their interface character with respect to deformation depends on the same few basic variables.

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## 1. Introduction

### 1.1. Processing–microstructure relationships

It is widely recognized that internal grain boundaries (homophase interfaces) and heterophase interfaces greatly affect the properties of polycrystalline metals [1–7]. Through the control and optimization of these internal interfaces, potentially superior metals with unprecedented strengths and robustness can be developed [4,5,8,9]. For instance, it has been shown that changes in interface properties within nanostructured metals can be made via different processing methods [10–16]. Thermodynamic, near-equilibrium processes, such as solid-state phase transformation, epitaxial growth or solidification processing, can produce highly textured nanostructured single-phase

and composite metals with interfaces that are structurally ordered at the atomic scale [17–21]. They adopt interfaces with nearly the same crystallographic character (narrowly distributed within 15°) throughout the material. By virtue of this near-structural perfection, these materials have shown outstanding thermal stability [22], radiation tolerance [23,24] and strength [25,26]. Far-from-equilibrium mechanical processing, such as severe plastic deformation (SPD) techniques [14,15,27–30], can also produce ultra-fine-grained and nanostructural metals comprised of a high density of interfaces. In single-phase SPD metals, while a small fraction of ordered boundaries has been reported [31], most of the grain boundaries are structurally disordered, and are often referred to as “non-equilibrium” boundaries [14,32,33]. Likewise, in bimetal wires fabricated by wire-drawing and bundling, several types of interfaces, both ordered and disordered, can form [29,34,35]. Although imperfect in structure, these SPD nanostructures possess superior strength, and in some cases, ductility

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[15,36,37]. However, the disorder renders them microstructurally unstable with respect to heating [38–41].

### 1.2. Observation of crystallographically stable interfaces

Recently, it was reported that a well-known SPD process, called accumulative roll bonding (ARB) [28,42–45], induced the formation of an ordered bimetal Cu–Nb interface that prevailed ubiquitously over the bulk two-phase Cu–Nb layered composite. After extreme strains and substantial layer refinement (99.96–99.99% rolling reduction and individual layer thicknesses of  $h = 200$ – $700$  nm), a predominant bimetal interface emerged with a crystallographic character that was narrowly distributed about  $\{112\}\langle 111\rangle\text{Cu}||\{112\}\langle 110\rangle\text{Nb}$  [44] and with a highly ordered atomic structure with little to no detectable defects [44,46,47]. In light of prior reports of interface structures after SPD, the emergence of a highly oriented and atomically ordered predominant interface from ARB is unexpected. The present knowledge base for microstructural evolution in SPD is insufficient for explaining SPD-induced ordering of interface structure.

Recently, two variables were proposed to influence the crystallographic stability of this interface [48]. The first variable is plastic stability in co-deformation, which refers to the preservation of interface character during plastic deformation. Crystal plasticity finite-element (CPFE) simulations of a Cu–Nb bicrystal with the  $\{112\}\langle 111\rangle\text{Cu}||\{112\}\langle 110\rangle\text{Nb}$  interface under plane strain compression found that its character misoriented only a few degrees to a stable end state [49,50]. Repeating the analysis for other bicrystals in single-crystalline or polycrystalline layers suggested that when an interface is composed of two stable rolling orientations, its character tends to be preserved in rolling [49,50]. This result forecasts that many interface characters could be plastically stable in co-deformation, not only the  $\{112\}\langle 111\rangle\text{Cu}||\{112\}\langle 110\rangle\text{Nb}$  interface. The second variable is interface formation energy, which can be calculated using molecular dynamics (MD) simulation [47,51]. As layers refine in the ARB process and the interface density increases, a lower formation energy interface would result in lower stored energy in the material. However, while the  $\{112\}\langle 111\rangle\text{Cu}||\{112\}\langle 110\rangle\text{Nb}$  interface does not have the highest formation energy ( $825\text{ mJ m}^{-2} < 1000\text{ mJ m}^{-2}$ ), it also does not correspond to the lowest one (Kurdjumov–Sachs or Nishiyama–Wasserman  $\sim 576$ – $586\text{ mJ m}^{-2}$ ). The conclusion was that both variables mattered and attaining an optimal combination of both presents a severe constraint that far fewer interfaces satisfy.

### 1.3. Objectives: role of deformation twinning

These ideas were developed assuming that both metals deform by slip only. In this work, we investigate the evolution of interfaces when the face centered cubic (fcc) Cu phase undergoes twinning in addition to slip. When  $h$  in

the Cu–Nb composite is refined via ARB to nanoscale dimensions ( $< 60$  nm), the Cu phase is found to deform via slip and twinning. Because of this change in deformation mechanism, the  $\{112\}\langle 111\rangle\text{Cu}||\{112\}\langle 110\rangle\text{Nb}$  interface becomes unstable. Here, with microstructural characterization and crystal plasticity analyses, we reveal that another stable, predominant interface emerges after twinning, distinct from the one that develops when the crystals deform purely by slip, which remarkably also exhibits a regular atomic structure. We analyze its origin and relationship to deformation twinning. The results enable us to advance the set of stability criteria not only to explain the development of both stable interfaces but also to provide insight into creating other crystallographically stable interfaces via deformation processing.

## 2. Observations of evolution in texture and interface character

### 2.1. ARB processing

ARB processing of the Cu–Nb layered composite begins with an alternating stack of 2 mm sheets of coarse-grained polycrystalline Cu and Nb in equal fractions [45,52]. Prior to ARB processing, the as-received Cu (99.99% purity) sheets were rolled to 60% reduction to 2 mm and subsequently annealed at  $450^\circ\text{C}$  for 1 h. The as-received Nb sheets (99.94% purity) were rolled to 30% reduction to 2 mm and annealed afterwards at  $950^\circ\text{C}$  for 1 h. To prevent exposure of the Cu–Nb interfaces to air during ARB processing, the stack was clad on the top and bottom by two half-thick layers of Cu (1 mm). Through repeated rolling, cutting and restacking, the process imposes radically extreme strains, achieving three to six orders of magnitude refinement in the individual layer thickness,  $h$ , from 2 mm to 10 nm [45,52]. As the layers are refined, they remain continuous and the bimetal interfaces remain chemically sharp due to the immiscibility of these two metals [46,53]. For  $h$  from  $45\text{ }\mu\text{m}$  to 200 nm, scanning electron microscopy (SEM) was used to gather statistical data on layer thickness [52]. For  $h$  from 135 to 10 nm, transmission electron microscopy (TEM) was used for this purpose [52]. The analyses showed that the average thickness of the Cu and Nb layers at each strain level corresponded well to the nominal value calculated from the applied rolling reduction. This indicates that as  $h$  is refined from  $45\text{ }\mu\text{m}$  to 10 nm, the two metals plastically co-deformed [54]. The above is a brief account of the materials and processing used and more information can be found in prior works [45,55].

### 2.2. Theoretical textures of monolithic Cu and monolithic Nb

The ARB process imposes the same deformation state as conventional rolling. It is, therefore, worthwhile to compare the textures of the individual phases within the composite with textures that develop when each phase is

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