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Structural observations of the interface of explosion-bonded Mo/Cu system

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The structure of the interface of explosion-bonded Mo/Cu system has been examined by optical and electron (TEM, AEM) microscopy. Molybdenum and copper can be directly bonded along the flat interface without a diffusion zone, but only on the copper side of the interface is a thin (about 10 μm thick) "bond layer," which consists of very fine subgrains with an ill-defined boundary. The microstructure in the bond layer is generated by severe dynamic deformation due to jetting and subsequent recovery with frictional heating. On the molybdenum side, originally-existing and elongated subgrains are observed just adjacent to the interface, even after bonding. These results indicate that jetting can occur only on the copper side, with a strength much lower than molybdenum because bonding must be carried out at an impact pressure as low as possible to avoid cracking of molybdenum as well as melting of copper during welding. *© 1998 Kluwer Academic Publishers*

1. Introduction

Explosion bonding is generally considered to be a cold-pressure welding, since the materials on both sides of a bond interface can be observed to be directly welded at optical microscopic magnifications. However, transmission electron microscopic (TEM) studies have shown, in most cases, that there is a very thin layer of submicroscopic structure, which has been severely deformed and subsequently recrystallized, or melted by frictional heating and rapidly solidified. So far, TEM studies were carried out mainly on simple systems of combinations between similar metals, or a metal and its alloy, such as Cu/Cu [1, 2], Cu/Cu-2% Be [3], Al-3% Cu/Al-3% Cu [4], and mild steels/stainless steels [5, 6]. This is due to difficulties in preparing TEM foils; the dissimilar metals on each side of a bond interface impede a uniform thinning rate in the electrolytic polishing procedure.

The explosion bonding is especially useful for welding dissimilar metals—with extremely different properties, such as melting point and mechanical strength—which cannot be welded by any other method. The importance of the structural examination of the bond interface lies in an understanding of the bonding mechanism and also of the composite properties such as bond strength, secondary formability, thermal-fatigue strength, etc. Nevertheless, TEM studies on the important systems of dissimilar metals with different properties have not been carried out except with a few systems of Al/Fe [7] and Fe/Ti [8]. Explosion-bonded Mo/Cu is an important composite as semiconductor heat sinks, high current-carrying contacts, or recently as the diverter for fusion reactors, since both metals have good thermal and electrical conductivities. This system is also an interesting object to bond, because both metals have extremely different melting points and no intermiscibility, even in liquid states. This is one of the most difficult combinations to weld by conventional methods, such as hot rolling and diffusion bonding.

The purpose of the present paper is to examine the structure just adjacent to the interface and to discuss the bonding mechanism. Described elsewhere is the bond strength of the composite, the severely deformed structure, and the deformation texture along the interface with association of jet flow.

2. Experimental methods

The materials used for explosion bonding were 0.5 mm thick plates of commercially pure copper, and of arc-melted and hot-rolled molybdenum. Explosion bonding was carried out under appropriate conditions by Asahi Chemical Industry Co. Ltd. Specimens for optical microscopy were etched by following regeants; 10% NaOH/10% K3 Fe(CN)6/80% H2O solution for molybdenum and 45% NH4 OH/9% H2O2/46% H2O solution for copper. Specimens for the TEM studies were prepared from slices with a size of 3 mm × 1 mm × 0.15 mm, cut parallel to the bonding direction by wire sawing. These slices were ground to a thickness of 40 μm by diamond slurry and finally polished by an ion–milling device operating at 5 kV and 1 mA, aided by occasionally chemical-etching only the molybdenum to avoid forming a step at the interface. The foils were examined by a JEM 1000 electron microscope operating at 750 kV. The chemical composition near the interface was analysed by an analytical electron microscope (AEM) JEM 2000 EX operating at 200 kV, using a beam diameter of 50 nm.

3. Results

Fig. 1 shows optical micrographs of an explosion-bonded interface. The morphology of an explosion-bonded interface is always flat under the present experimental condition. On the side of the copper plate, the structure of extremely elongated grains observed in many other systems is not found along the interface, but in its vicinity grain boundaries become ill-defined and some features of plastic flow are discernible in the bonding direction (Fig. 1a). On the molybdenum side, many stripes of grain boundary run parallel to the interface. Ion etching revealed a thin layer of approximately constant thickness of 10 μm along the interface (Fig. 1c). The layer, later referred to as "bond layer," is easier to etch than molybdenum and bulk copper. There exist a number of faint streaks normal to the interface, which make the bond layer columnar-grain-like. Fig. 2 shows the microstructures of molybdenum immediately adjacent to and about 100 μm distant from the interface. The subgrains elongated parallel to the interface are the structure induced by hot rolling to prepare the plate for bonding. A high density of dislocations ($5 \times 10^{10}/\mathrm{cm}^2$) generated during bonding are homogeneously distributed in the subgrains—as shown in the inset—over the thickness of the plate though there is a slight tendency to recovery near the interface. Chemical composition across the interface was found by AEM to change abruptly from molybdenum to copper without a diffusion zone. Figs 3 and 4 show the general structural features and two typical microstructures in the bond layer. This layer is found to mostly be formed by very fine, recovered sub-grains of copper, while abnormally coarsened grains are occasionally observed along the interface (Fig. 3). The morphology of subgrains is elongated away from the interface (Fig. 4a) but always equiaxed within about 2 μm from it (Fig. 4b). The latter layer is a structure produced by being disturbed by friction at the interface. Fig. 5 shows the structures of bulk copper about 60 μm distant from the interface and bounded by the bond layer. In these regions a high density of dislocations are distributed very homogeneously and too densely to be distinguished separately, whereas further away from the interface, the dislocation density considerably decreases. (The cell structure will be reported in detail elsewhere.) Fig. 6 shows an over-width view of the bond layer, the width of which happens to be narrower than elsewhere. This indicates that the bond layer is easier to ion-etch than is bulk copper (Fig. 1c). Furthermore, the fine recovered structure has already developed into only two coarsened grains, extending over 10 μm along the interface.

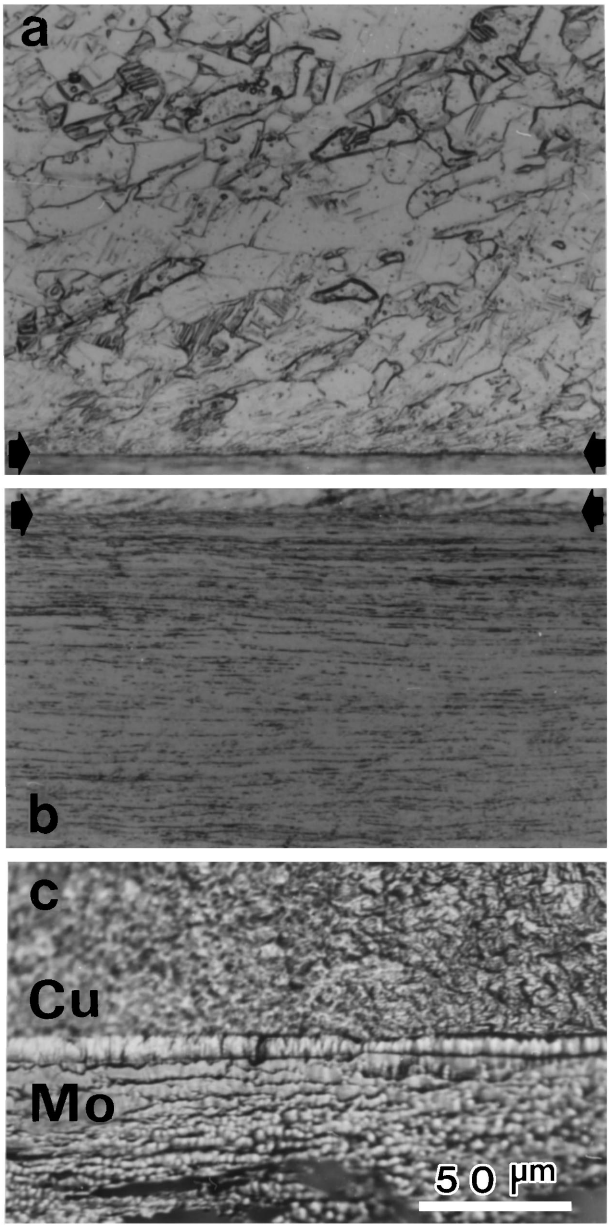

Figure 1 Optical micrographs of cross sections near the interface (arrows) of explosion-bonded Cu/Mo; chemically etched (a) copper and (b) molybdenum, and (c) ion-etched section. The explosion-bonding direction is horizontal.

4. Discussion

The structure of an explosion-bonded interface is basically produced by a severe deformation of materials, due to the viscous flow as jets, and is always influenced by a localized temperature rise with frictional heating. In a combination of similar metals, the structure is mainly controlled by the collision parameters, such as plate velocity (V_p), collision point velocity (V_c), and collision angle (θ) (Fig. 7), but in a combination of dissimilar metals, material parameters also influence the interface structure. These points will be briefly considered below.

4.1. Jetting condition for bonding dissimilar metals and the interface morphology

In many studies on the jetting phenomenon, bondings were carried out at an excessive energy level to remove difficulties in experiments [10], and there were usually relatively thick layers of coarse solidified structure along the interface. Under such a condition jetting is known to occur simultaniously on both sides of the interface, even in combinations of dissimilar metals

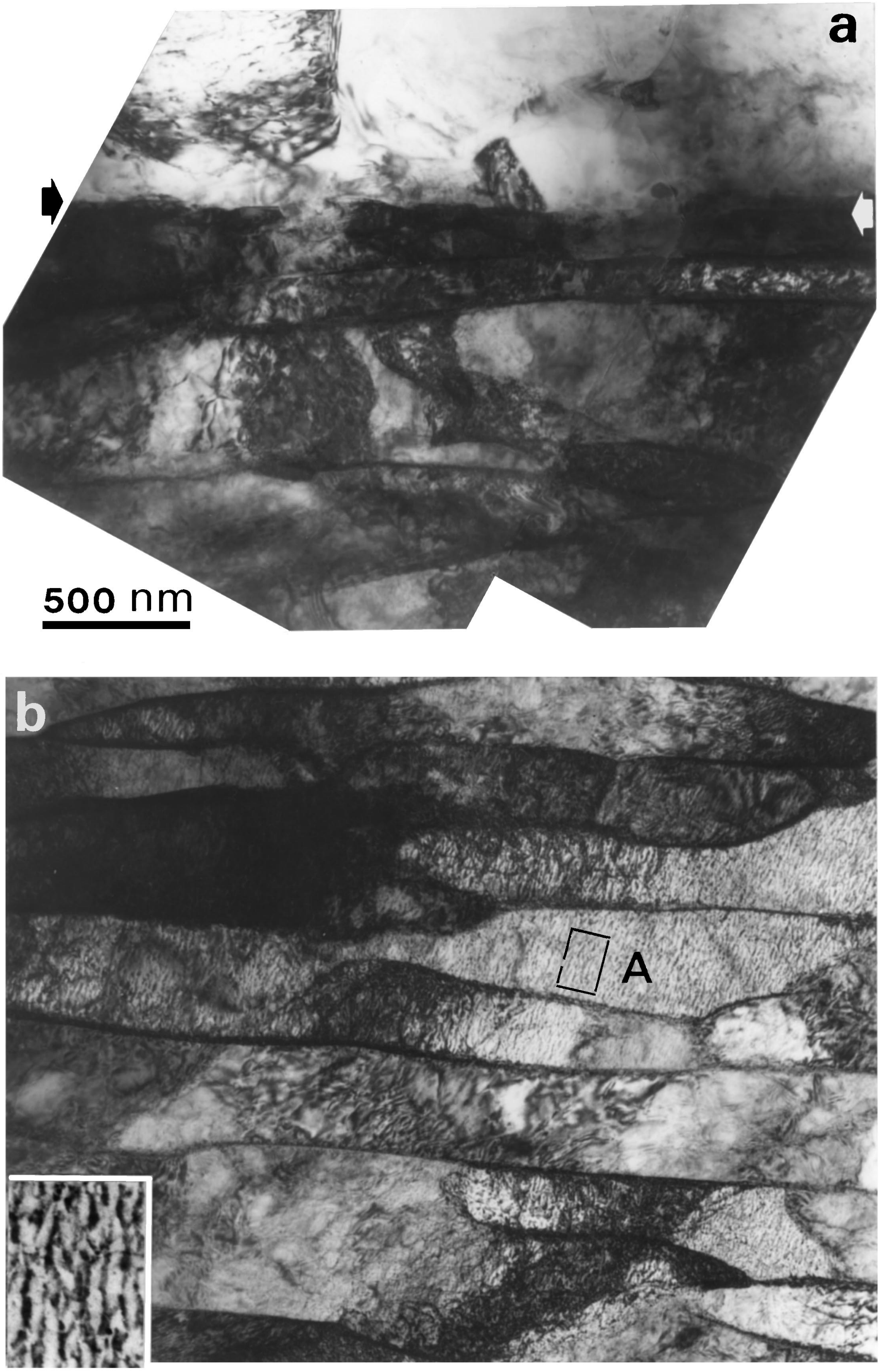

Figure 2 TEM photographs of molybdenum, (a) directly adjacent to and (b) about 100 μm distant from the interface (arrows). Region (A) is magnified in the inset.

with very different properties [11]. In practice, however, bonding must be done by using a sufficient but not excessive energy level to avoid detrimental structures, such as the formation of intermetallic compounds and a coarse solidified structure. Molybdenum and copper have very different melting temperatures and elastic properties as shown in Table I. For such a combination of dissimilar metals with very different properties, structural observations of the explosion-bonded interface have not been carried out except in a few combinations [7, 8]. Here, the aspects peculiar to such a system will be discussed on the base of the near-interface

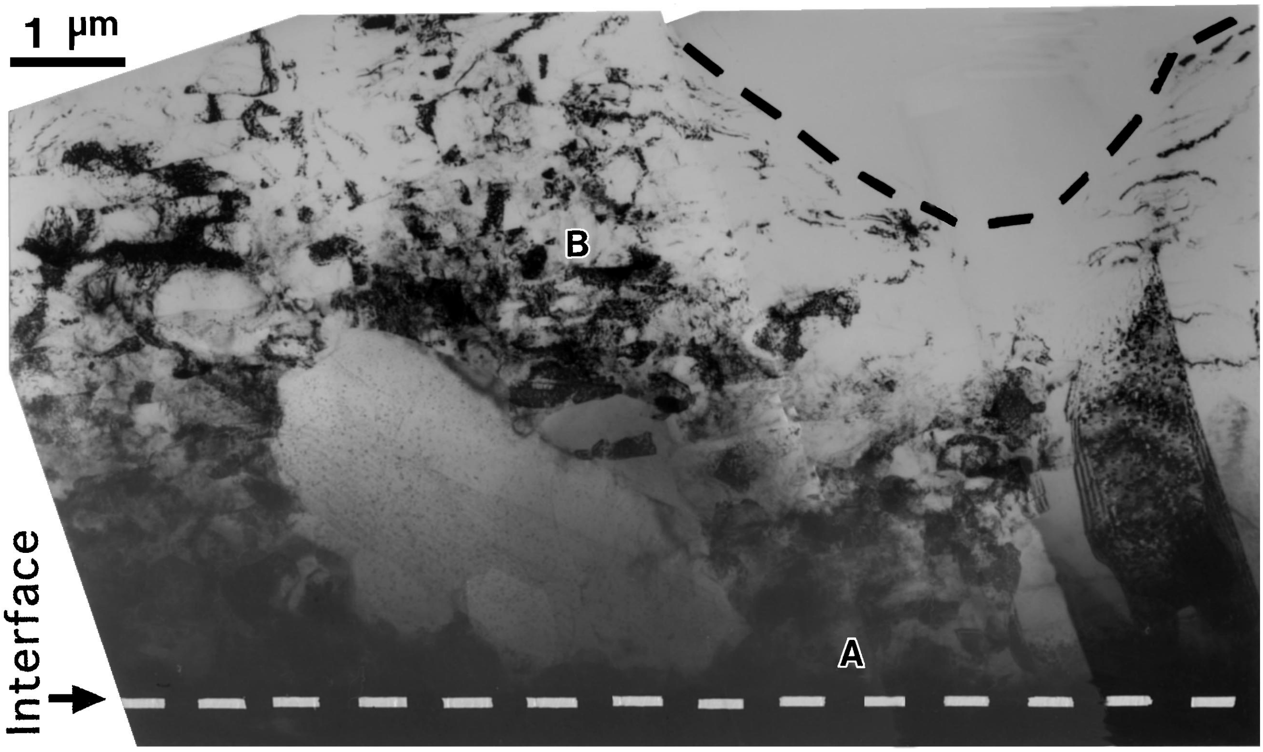

Figure 3 The general feature of the structure in the bond layer.

TABLE I Comparison of melting points, bulk moduli and critical hydro-pressures for dynamic deformation.

	Melting point (K)	Bulk modulus (G Pa)	Critical hydro-pressure (G Pa)
Molybdenum	2888	288	7.7
Copper	1356	92	2.5

structure, which is closely associated with the jetting phenomena.

When molybdenum and copper collide at a relative velocity of V_p, the impact pressure (P) at the collision point (C) is given by [12]

$$P = P_a = P_b = \rho_a V_p U_a / [1 + \rho_a U_a / \rho_b U_b]$$

where ρ and U are the density and the velocity of sound in each metal (the subscripts a and b refer to molybdenum and copper respectively). On the other hand, the critical hydropressure (P_c) for dynamic deformation of a metal is expressed by $P_c = 0.027$ K, where K is the bulk modulus [13, 14]. The critical hydropressure of both metals are very different (Table I), but only the same value of impact pressure can be produced on each side of the interface. In the present experiment a high density of dislocations were observed all over the thickness of molybdenum and copper. This suggests that the impact pressure could reach a value greater than those in Table I and that the deformation occurred by its propagation through the thickness. By the way, jetting can occur only when the impact pressure reaches a value many times higher than the critical hydropressure and when the metal can be transiently treated as a fluid. However, molybdenum is one of the rather brittle metals at room temperature, and so the impact pressure had to be minimized in order to avoid cracking it as well as melting the copper during welding. The bond layer, which is produced by jetting—as briefly described below—was observed only on the copper side. This indicates that the jetting condition was met only for copper, but insufficiently for molybdenum. Fig. 8 shows a plan view of copper facing the interface, from which molybdenum was etched away. The pattern of striations in the direction of explosion bonding is thought to be transferred from the roughness on the molybdenum facing the interface because it is similar to the chemically etched surface of molybdenum plate before welding (Fig. 8c). The roughness on molybdenum was produced at the collision point during welding, since the deformation anisotropy with the orientation of elongated subgrain emerged due to an impact pressure level insufficient for the viscous flow as a jet. So far, anisotropic behavior caused by the insufficient fluidity of the material near the interface has been reported [1]; in a Cu/Cu system, explosion-bonded at a just-necessary level of impact pressure, the interface may be irregularly shaped or regularly wavy only by changing the grain size. As discussed above, jetting can occur only on the copper side in the present experiment. Finally, in this case surface film and contaminants on molybdenum can be scraped off by copper jet, while a clean surface of copper is obtained by jetting off surface film itself.

Although a wavy interface is generally desirable for achieving a high bond strength, the flat interface was obtained in the present experiment. According to Cowan et al., the interface morphology depends on a collision-point velocity (V_c) and a flat interface can be obtained at a low velocity [15]. In this experiment the bonding had to be carried out at an impact pressure or plate velocity

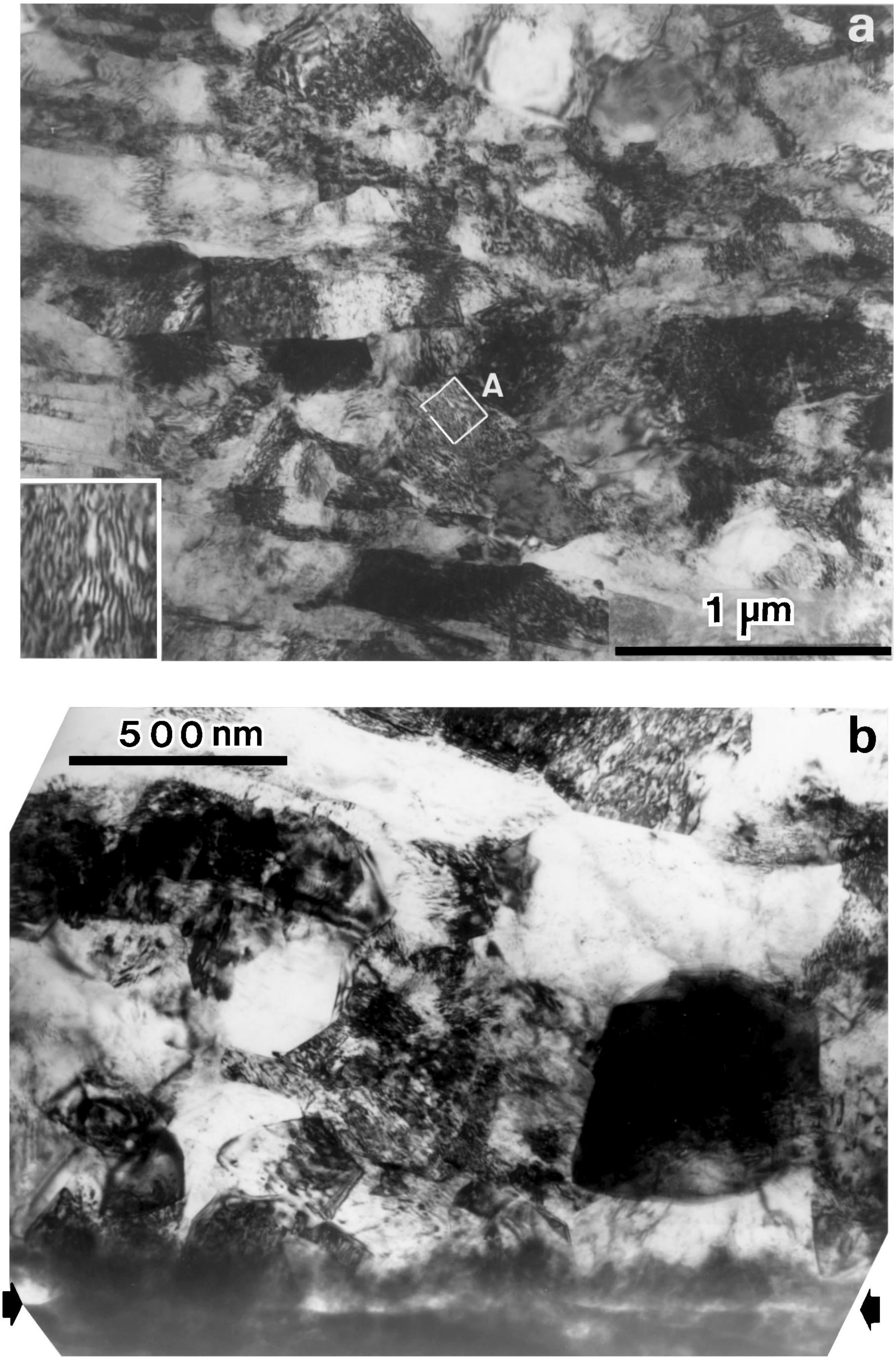

Figure 4 Two typical microstructures in the bond layer; (a) away from, and (b) directly adjacent to the interface (arrows), which correspond to the regions (B) and (A) in Fig. 3, respectively.

(V_p) as low as possible to avoid cracking of the molybdenum. This led to a lower collision-point velocity at a fixed collision angle (θ)—because $V_c = V_p \sin\theta$ for a small θ—and produced the flat interface. It should be stated in this connection that the bonding was done just under the condition where the interface morphology

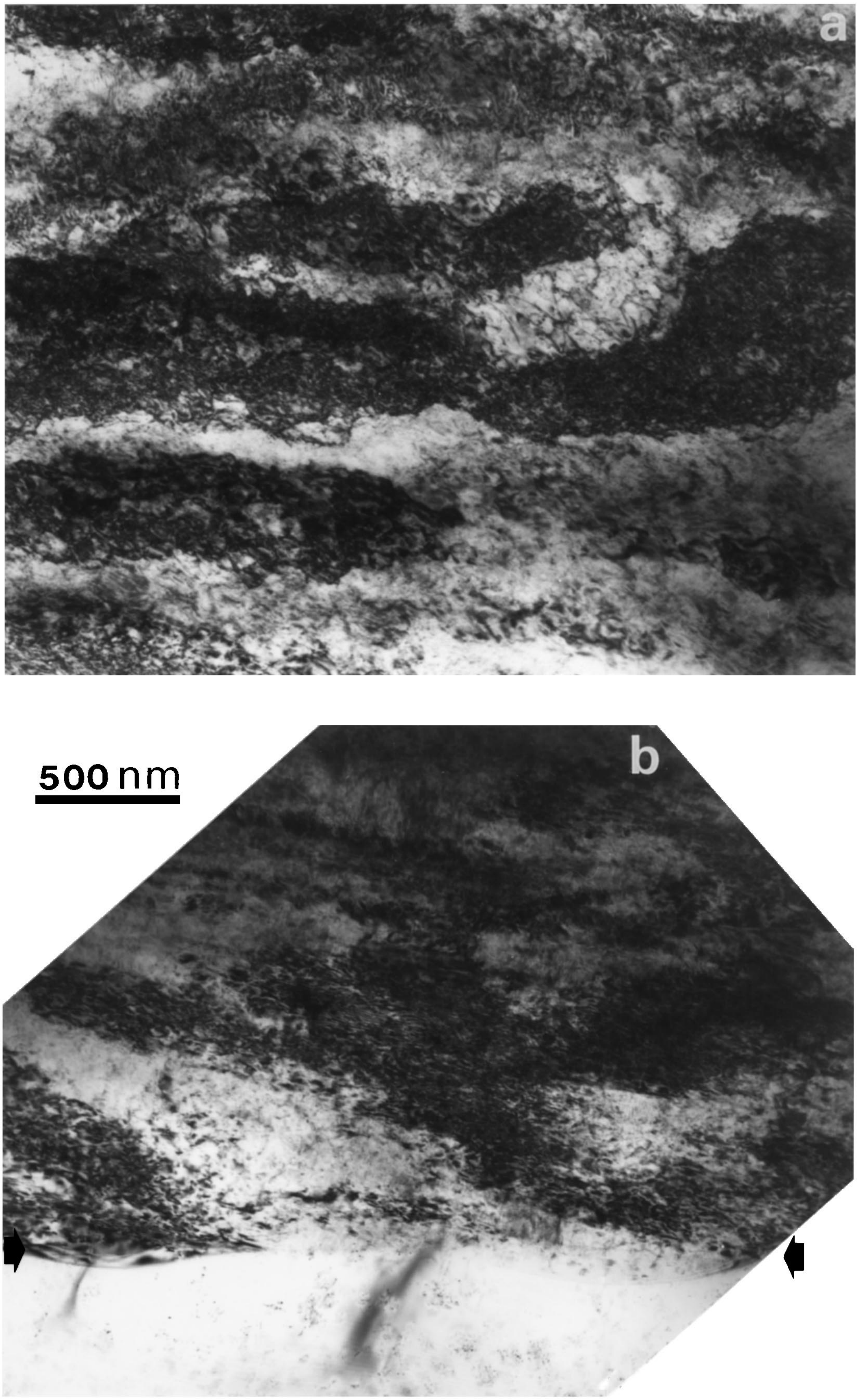

Figure 5 Structures of bulk copper; (a) about 60 μm distant from the interface and (b) bounded by the bond layer. Arrows indicate the boundary between the bond layer and bulk copper.

changed from flat to wavy, as shown by the fact that the interface is dotted with patches of ripples of very small amplitude, up to 10 μm deep in the plan view of the interface as indicated in Fig. 8b.

4.2. Microstructures along the interface

Although a high density of dislocations were observed all over the thickness of both plates, severe deformation by the forward viscous flow as a jet occurred near

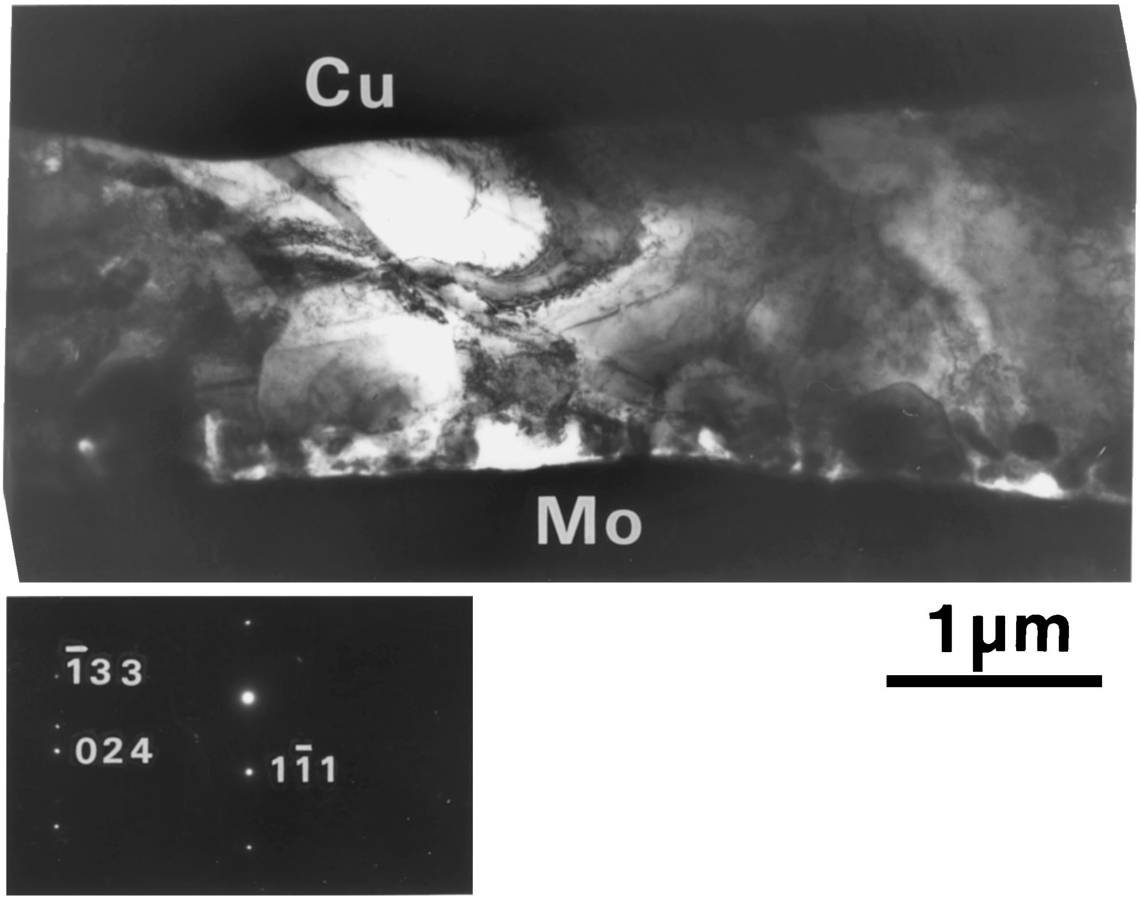

Figure 6 The structure and diffraction pattern of a bond layer whose width happened to be narrower than elsewhere.

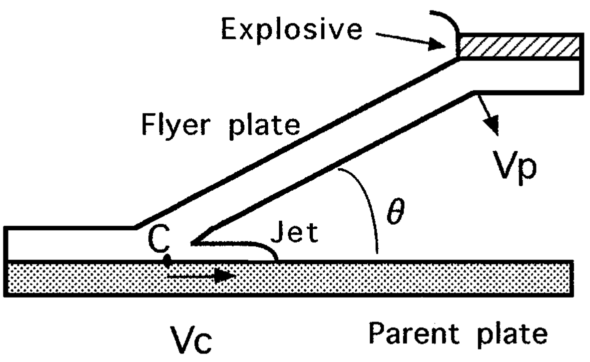

Figure 7 Shematic of an oblique collision showing formation of a jet.

the interface, only on the copper side, especially in the bond layer (Figs 3 and 4). The severe deformation in the bond layer developed a deformation texture with a specific orientation different from that of bulk copper [16]. This is responsible for the difference of ion-etching rate between them (Figs 1 and 6) because it is known to be dependent on crystal orientation [17]. The microstructure in the bond layer was generated by such deformation and subsequent recovery with frictional heating, and is characterized by ill-defined subgrain boundaries (Fig. 4a). Such a structural feature is caused partly by the superimposition of more than two grains through the foil thickness, as evidenced by the moire pattern in the inset (Fig. 4a). However, the peculiar structure, which has not been observed in any conventionally static-deformed and recovered material, essentially resulted from the dynamic deformed structure where dislocations were distributed very densely and homogeneously (Fig. 5a). Subgrains arising from the dynamic deformed structure have small misorientations between them. This is responsible for the ill definition of subgrain boundary. Such structure is not usually recrystallized into small grains but into extremely large ones, as occasionally observed along the interface (Figs. 3 and 6). Such anomalous coarsening behavior appears to be common to dynamic deformed structure because it has been observed in shock-compressed and annealed nickel [18], or along the interface of explosion-bonded Cu/Cu [1] and mild stainless steel [5].

In addition to the large difference in mechanical strength, the Mo/Cu system is characterized by no intermiscibility and no formation of intermetallic compounds, and these properties can influence the interface structure and its thermal stability. In the field of solid friction [19], it is known that the friction, and then frictional heating, between dissimilar metals is very dependent on the extent of their intermiscibility; the friction coefficient (μ) is 4 for a mutually soluble couple, Cu/Ni, but 1.4 for Cu/W with no intermiscibility, which is similar to the Mo/Cu. Therefore, it appears that a transiently melted layer along the interface is rather unlikely to occur with other fixed welding parameters in the present system because of a small amount of frictional heating and rapid diffusion of heat out of the interface, due to high thermal conductivities of both metals. In fact, the melted zone was not observed here, though a friction-disturbed layer always existed directly adjacent to the

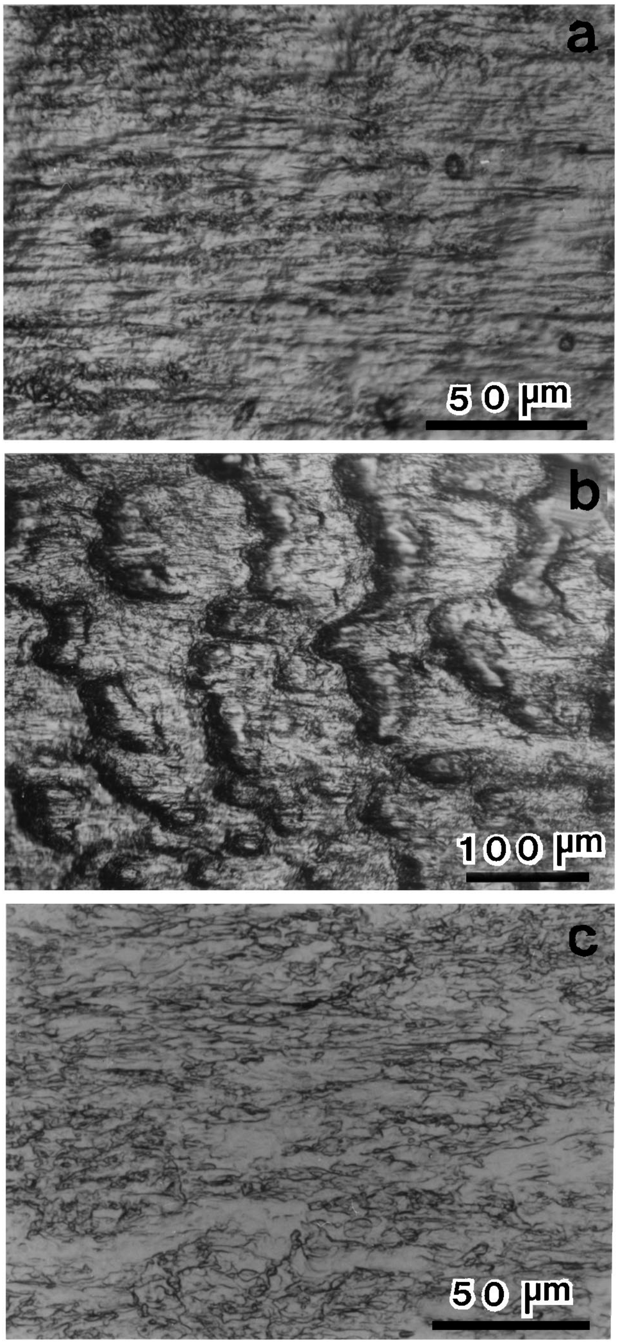

Figure 8 Plan views of copper surface facing the molybdenum which had been etched away after bonding ((a), (b)) and chemically etched surface of raw molybdenum plate before bonding (c).

interface (Fig. 4b). On the other hand, a very thin (less than 2 μm) but melted layer directly bounded by the severely deformed structure was formed both in Al/Fe [7] and Al/Ti [8] systems, which are limited examples of structural examination on the explosion bonded interface between dissimilar metals where the intermetallic compounds occur in equilibrium. The thermal stability of the Mo/Cu composite is an important issue because it is subjected to cyclic heating in its applications, such as semiconductor heat sinks and high current-carrying contacts. In general, a good bond strength in the as-welded state, e.g., in the Al/Fe system [7], becomes extremely deteriorated by heating, due to the formation of brittle intermetallic compounds. There is no such a defect in the Mo/Cu composite, but the coarsening behavior of the dynamic-deformed structure on the copper side is very different from that of the static deformed one, as described above, and this effect on the thermal fatigue strength remains to be determined.

5. Conclusions

Structural examinations of the interface of explosion-bonded Mo/Cu system have been carried out by optical and electron (TEM, AEM) microscopy.

(1) Molybdenum and copper can be directly bonded along the flat interface without a diffusion zone.

(2) Only on the copper side of the interface is found a thin (about 10 μm thick) "bond layer," which consists of very fine subgrains with an ill-defined boundary. The microstructure in the bond layer is generated by severe dynamic deformation, due to the forward viscous flow as a jet and the subsequent recovery with frictional heating.

(3) On the molybdenum side, however, originally-existing and elongated subgrains are observed near the interface as well as far away from it.

(4) In this system, between constituent metals with very different properties such as mechanical strength and melting point, bonding must be carried out at an impact pressure as low as possible to avoid cracking of molybdenum as well as melting of copper during welding. In this case, jetting can occur only on the copper side with a strength much lower than molybdenum because the jetting condition is met only for the copper side.

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