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Some Comments on "The Effects of Various Properties of FCC Metals on Their Adhesion as Studied With LEED"

T. EDMONDS, J. J. MCCARROLL and R. C. PITKETHLY

The British Petroleum Company Limited

BP Research Center

Sunbury-on-Thames

Middlesex, England

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In the above paper¹ D. H. Buckley has reported some LEED investigations of adhesive contact between metal surfaces. We believe that misinterpretation of part of the data may have led Buckley to some erroneous conclusions concerning the basic processes involved in adhesion. Our comments are as follows.

1. Orientation Effects: the Adhesion of Gold to the (111), (100) and (110) Planes of Copper

The LEED evidence presented relates to the (111) and (100) faces. The results on both faces are essentially similar. They do not however show that epitaxy has occurred. The formation of an epitaxed layer of gold on copper (111) would result in the diffraction pattern drawn schematically in Figure 1a, neglecting any strain in an epitaxed gold layer and/or the formation of any coincidence nets. Epitaxy would not lead to splitting of the (0,0) spot, apparently shown by Buckley in his Figure 2e. The formation of angled bicrystals of copper, induced by the strain of adhesive contact, could result in the diffraction pattern shown schematically in Figure 1b—which closely resembles Buckley's diffraction photographs in Figure 2. An exactly similar picture emerges from the results presented for the (100) face of copper.

We would thus propose that, although Buckley has detected the presence of gold on the copper after adhesive contact, the gold has not formed an epitaxed layer nor any ordered structure on the (111) and (100) faces of copper. The only effect observed is strain induced surface bicrystallinity.

An alternative explanation to that of strain-induced bicrystallinity can be advanced in terms of Rhead's work² on vicinal surfaces which result from surfaces cut at a small angle to a low index face. Insufficient data are available in Buckley's paper to confirm or deny this hypothesis and we consider that strain-induced bicrystallinity is the best explanation to account for Buckley's diffraction photographs.

We suspect that a similar situation exists for the (110) face of copper, the behaviour of which, though not described by Buckley, is stated to be similar to that of the (111) and (100) faces. Figure 10b in Buckley's paper can also be interpreted to yield a similar explanation for the contact of copper to the (111) face of nickel.

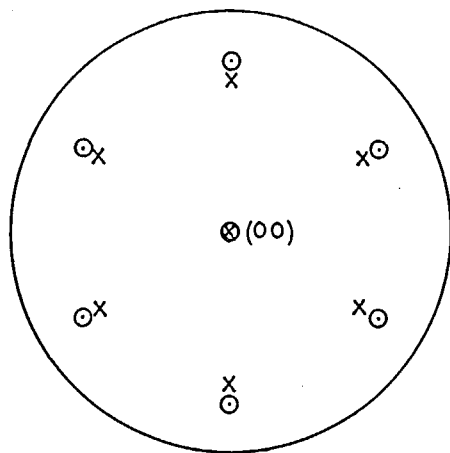


Figure 1 a. Schematic reciprocal net for a thin epitaxial (III) layer of gold on the (III) plane of copper.

⊗ Reciprocal net points of (III) plane of copper.
 X Reciprocal net points of epitaxial (III) plane of gold.

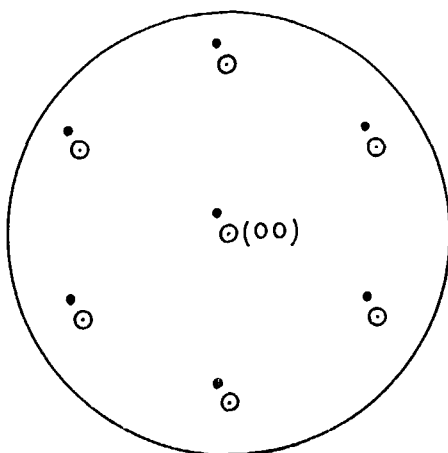


Figure 1 b. Schematic of two copper (III) planes mutually set at a small angle.

⊙ Reciprocal net points of first (III) plane of copper.
 ● Reciprocal net points from tilted (III) plane of copper.

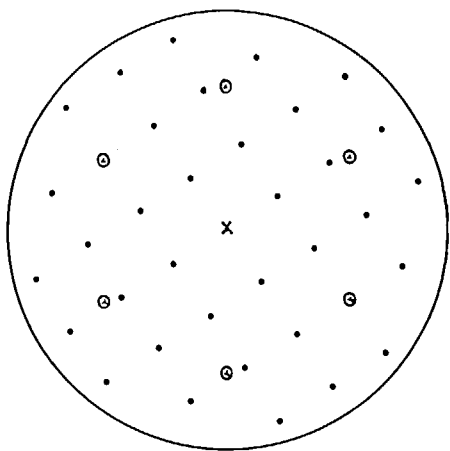


Figure 2 a. Schematic reciprocal net of one domain of a reorientated Ni(100) (2 × 2) - S layer on Ni (III).

⊙ Reciprocal net points of (III) plane of nickel.
 ● Reciprocal net points of Ni (100) (2 × 2) - S layer. For dimensions see ref. 5.

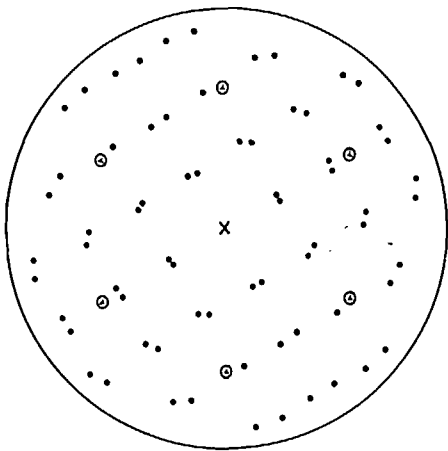


Figure 2 b. Schematic of 2 mirror image domains of a reorientated Ni(100) (2 × 2) - S layer on Ni (III).

2. Alloying

The diffraction photographs (Figure 6) presented by Buckley for the contact of Au(111) against an alloy Cu-Al(111) face also appear to show a similar effect to that discussed above—strain induced bicrystallinity. Figures 6b and 6c closely resemble our Figure 1b.

3. The Adhesion of Lead to the (111) Face of Nickel

• Figure 10c shows the LEED pattern resulting from the adhesion of lead to Ni(111). Similar patterns are shown elsewhere by Buckley for the contact of quartz³ and tungsten oxide⁴ with Ni(111). In one case (quartz) the pattern is described as arising from a distorted Ni(111)(2 x 2), with additional diffraction features and in the other (tungsten oxide) as being from a Ni(111) (3 x 3) R30° structure. The lead/Ni(111) case described here appears to be considered as epitaxy.

In our opinion none of these three explanations is correct. The diffraction features observed in this case can arise from the formation of a reorientated layer of Ni(100) (2 x 2)-S which we have described elsewhere⁵. One domain is shown schematically in Figure 2a and two mirror image domains in Figure 2b. The single domain is what was found in the quartz and tungsten oxide cases, the two mirror image domains being found with lead. Our assignation of the structures witnessed by Buckley to sulphur is based on the relative intensities of the spots arising from the reorientated layer, particularly the (0, $\frac{1}{2}$) spots, which are relatively quite intense. When the structure is formed with carbon compounds the relative intensities of the (0, $\frac{1}{2}$) spots is much lower.

The question must then be asked—where did the sulphur come from? The most likely interpretation is that the sulphur arose from the nickel crystal itself. Energy released by the adhesion processes could have caused diffusion of sulphur to the surface to form the reorientated layer. In view of the very brief clean up procedures used by Buckley (15 min at 10⁻⁶ torr of O₂) compared to those found necessary by ourselves⁶ this would not seem unreasonable. Sulphur is known to be an impurity which can diffuse to the surface of nickel⁷.

The appearance of single domains or two mirror image domains is also of interest. It may imply that the adhesive contact method employed by Buckley involved directional shear forces.

Conclusions

We would thus suggest that in the following systems

- (a) gold/copper
- (b) gold/copper-aluminium alloy
- (c) copper/nickel
- (d) lead/nickel

the evidence presented suggests no epitaxy but in cases (a), (b) and (c) strain induced bicrystallinity and in case (d) the diffusion of impurity atoms to the surface of the nickel.

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