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MICROSTRUCTURE OF Y2O3. FLUXED HOT-PRESSED SILICON NITRIDE

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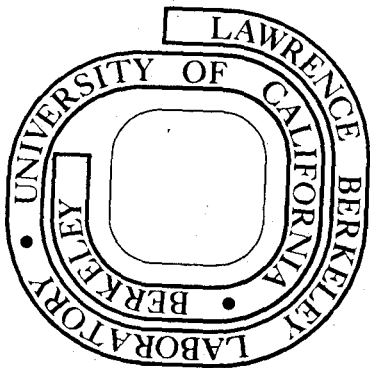
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MICROSTRUCTURE OF  $Y_2O_3$  FLUXED HOT-PRESSED SILICON NITRIDE

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ABSTRACT

Detailed microstructural analysis of a 10 m/o  $Y_2O_3$  fluxed hot-pressed silicon nitride reveals that in addition to the yttrium-silicon oxynitride phase located at the multiple  $Si_3N_4$  grain junctions there is a thin boundary phase 10-80Å wide separating the silicon nitride and the oxynitride grains. Also x-ray microanalysis from regions as small as 200Å across demonstrates that the yttrium-silicon oxynitride,  $Y_2Si[Si_2O_3N_4]$  phase can accommodate appreciable quantities of Ti, W, Fe, Ni, Co, Ca, Mg, Al and Zn in solid solution. This finding taken with the observations of highly prismatic  $Si_3N_4$  grains enveloped by  $Y_2Si[Si_2O_3N_4]$  suggest that densification occurred by a liquid phase "solution-precipitation" process.

## I. Introduction

The high temperature loss of strength exhibited by MgO hot-pressed silicon nitride is generally attributed to the presence of a vitreous silicate inter-granular phase having a low softening temperature<sup>(1,2)</sup>. The silicate, formed from the surface silica present on the silicon nitride powders and the magnesium oxide, melts at 1500-1600°C and acts as the densifying flux in liquid phase sintering. An important role is also played by the silicate in concentrating alkali impurities, such as calcium, in solution; the softening temperature of the silicate is further lowered, especially by the calcium oxide impurity<sup>(3)</sup> causing the premature loss of strength.

After the detrimental effect of the silicate had been identified, a number of attempts were made to improve the high temperature properties of silicon nitride. Basically they are of two types<sup>(4)</sup>. Firstly, the elimination of the glassy silicate phase both by decreasing the alkali impurities in the starting  $\alpha$ -silicon nitride powders and by minimising the amount of MgO additive. Secondly, promoting the formation of a more refractory intergranular phase by using alternative additives such as  $Y_2O_3$  and  $Ce_2O_3$ . The use of higher purity starting powders has yielded a steady improvement in the high temperature properties, but the most radical improvement was made by using yttria as a fluxing aid in place of magnesia<sup>(5,6)</sup>. Interestingly this was done with relatively impure  $\alpha$ -silicon nitride powders and the highest strength materials were then obtained with appreciable quantities of yttria, considerably in excess of that required merely to react with the surface silica. Subsequent investigations by Rae et al.<sup>(7)</sup> showed that the effectiveness of yttria as an additive in hot pressing silicon nitride is primarily due to the formation of an

yttrium-silicon oxynitride,  $Y_2Si[Si_2O_3N_4]$ , stable to at least  $1600^\circ C$  in an inert atmosphere. They also suggested<sup>(7)</sup> that, because of its structural similarity with the melilite silicates, it was capable of dissolving many of the impurities that formed a glass when MgO was used as an additive.

Although the use of  $Y_2O_3$  has clearly led to improved high temperature properties, the material still exhibits a decrease in strength at high temperatures, albeit higher than when MgO is used. Since this decrease occurs below the temperature at which the yttrium-silicon oxynitride itself becomes unstable, a number of questions about the role of the yttria remain.

This research was undertaken to determine whether the yttrium and impurity elements are solely in solution in the yttrium-silicon oxynitride, as suggested by Rae et al., and whether an intergranular phase, which may be responsible for the loss of strength, is present in addition to the crystalline yttrium-silicon oxynitride phase. The presence of a thin intergranular film was investigated using the electron microscopy technique of lattice fringe imaging used previously<sup>(8,9)</sup> to study the second phase in MgO hot-pressed silicon nitride. The spatial distribution of the elements in the microstructure was determined by x-ray micro-analysis in the electron microscope.

## II. Experimental Details

The silicon nitride sample, kindly supplied by Dr. F. F. Lange, was prepared, as part of the phase equilibrium study, by hot-pressing  $\alpha-Si_3N_4$  powder with 10 m/o  $Y_2O_3$  at  $1780^\circ C$  and subsequent cooling to room temperature.

Foils for electron microscopy were prepared from 0.2mm thick slices cut using a diamond wheel. The slices were then hand ground on silicon

carbide papers until they were  $<50\mu\text{m}$  thick, diamond polished and finally thinned to electron transparency using 5kV argon ion beams incident on the surface at  $15^\circ$ . Before examination, the thinned foils were coated with a thin layer of evaporated carbon to avoid surface charging in the beam of the electron microscope.

The lattice fringe images were taken with a Philips EM301 electron microscope fitted with a high resolution stage. The technique, by which the lattice planes on either side of the grain boundary are imaged simultaneously, has been described in detail previously<sup>(9)</sup>. The x-ray micro-analysis was performed using another EM301 electron microscope fitted with a scanning and EDAX attachment. The advantages and disadvantages of this technique have been described elsewhere<sup>(10)</sup>. By using scanning transmission microscopy (STEM) electron optics x-ray micro-analysis could be obtained with a spatial resolution of better than  $200\text{\AA}$  in favourable circumstances<sup>(15)</sup>. The principal disadvantage as far as this work is concerned was the inability to detect elements of atomic number smaller than twelve, with the instrument used. This limitation was due to the presence of a protective Beryllium window in front of the x-ray detector. One novel feature of the STEM micro-analysis attachment was the ability to obtain electron diffraction patterns from regions as small as  $30\text{\AA}$  across<sup>(11)</sup>.

### III. Microstructural Analysis

The microstructure of the  $\text{Y}_2\text{O}_3$  hot pressed silicon nitride when examined at low magnification in the electron microscope is seen to consist of two phases. The major phase is silicon nitride and has

a relatively equi-axed grain morphology. The minor phase is generally seen at multiple grain junctions as in figure 1a, but is also seen on occasions to be partially enveloping a silicon nitride grain as in figure 1b. In all cases, as in the micrographs of figure 1, the minor phase appears very much darker than the adjacent grains and remains so even on tilting. This lack of diffraction contrast, together with the observation by stereo-microscopy that the regions of minor phase were commensurate in thickness to the adjacent silicon nitride grains, indicates that the yttrium, an element with a much higher electron scattering factor, is concentrated in these regions and was subsequently confirmed by X ray micro-analysis.

Performing the chemical analysis it became clear that there are in fact two types of minor phase present. The first type is that illustrated in figure 1a, being concentrated at the multiple grain junctions. These were found to contain only silicon and yttrium by X ray micro-analysis, as in the trace A of figure 2a. Although no attempt was made to carry out quantitative X ray micro-analysis due to the irregular shape of the second phase particles and the unknown thickness of the foil, an approximate correction of the peak heights following the procedure described by Lorimer <sup>(12)</sup> indicates that silicon and yttrium are present in the ratio of 3:2.

The second type of minor phase differs from the first type in its chemical composition. In addition to containing silicon and yttrium these regions contain substantial proportions of impurity elements, notably transition and alkali elements. An example of the chemical constituents detected in one such region is shown for comparison



0 0 0 0 4 7 1 1 5 6 2  
0 0 0 0 4 7 0 1 7 6 6

-5-

by the curve B of figure 2a. (The central portion of the curve B is expanded and labelled in figure 2b). A striking feature of this second type of minor phase is that in each grain the proportions of all the elements are different, and in some of the larger grains the composition varies within the grain itself. Interestingly, the ratio of yttrium to silicon may vary from one grain to another, indicating that the impurity elements replace the silicon and yttrium in the phase. Whilst quantitative analysis of this could not be performed the effect was clear from the relative peak heights. On the basis of the X ray micro-analysis it becomes evident that the second type of minor phase is also morphologically distinct, since these are generally the minor phases that appear to envelope, partially or completely, a silicon nitride grain or a group of silicon nitride grains, as in figure 1b.

Some difficulty was experienced in determining whether the minor phases, particularly those at the three grain junctions, were crystalline or not, on account of their poor electron transparency. However, in the very thinnest areas, such as at the edge of the foils, electron diffraction patterns could be obtained using the technique of rocking beam micro-diffraction (for the very small phases), or standard selected area diffraction for the larger phase regions. The spot spacings found were found to correspond closely to those reported for

the yttrium-silicon oxynitride (7,13,14). Some of the spacings were also confirmed by subsequent lattice fringe imaging, using the fringe spacings in and adjacent silicon nitride grains as a precision scale.

The distribution of yttrium between the phases was also determined by X-ray micro-analysis. These observations showed that, within the sensitivity of the technique, there was no bulk solubility of yttrium in the silicon nitride grains. This agrees with the finding of Rae et al. that the lattice parameter of the silicon nitride remained unchanged after hot pressing with yttria. Furthermore, no segregation to the grain boundary of yttrium within the silicon nitride grains could be detected even using an analysis probe giving a pre-calibrated spatial resolution of  $180 \text{ \AA}$  (15).

At high magnifications a rather featureless phase between the silicon nitride and the yttrium-silicon oxynitride grains can be discerned in the very thinnest sections and appears as a bright line at the arrows in figure 3. From such micrographs this narrow region has a width measured in the range of  $10\text{-}80 \text{ \AA}$ . The fact that it is indeed a distinct phase was confirmed by lattice fringe imaging, as shown in figure 4. Here lattice planes in adjacent silicon nitride and yttrium-silicon oxynitride grains are imaged simultaneously with any spatial discontinuity between the set of fringes being identified as the location of a separate inter-granular phase. Using the known  $(10\bar{1}0)$  fringe spacing in the silicon nitride as a scale, the  $(21\bar{3}0)$  spacing in the oxynitride was measured to be  $3.43 \text{ \AA}$  which is very close to the value measured by X-ray diffraction ( $3.41 \text{ \AA}$ ). The width of the inter-phase film is  $26 \text{ \AA}$  at A.

The characteristics of the inter-phase phase are quite different to those of the intergranular phase in the MgO hot pressed silicon nitride (HS-130), as reported previously <sup>(9)</sup>. Firstly, the phase is far more common in the Y<sub>2</sub>O<sub>3</sub> hot pressed material and is found to be present at the interphase boundaries in the very thin areas of the sample. Secondly, no Si<sub>3</sub>N<sub>4</sub>-Y<sub>2</sub>Si[Si<sub>2</sub>O<sub>3</sub>N<sub>4</sub>] interphase boundary could be found where there was no boundary phase, whereas this was the most common finding in the MgO hot pressed silicon nitride. Thirdly, observations by both high magnification bright-field imaging and lattice fringe imaging indicate that the intergranular films are of approximately constant thickness along the interphase boundaries, as seen for instance in figures 3 and 4, whereas this was a most unusual finding in the MgO doped silicon nitride.

Attempts to determine unambiguously whether this interphase film is actually crystalline or non-crystalline were not successful. However, as they exhibited no diffraction contrast on tilting and did not produce any fringe images it is suggested that they are probably non-crystalline.

In addition to the above, a number of observations pertinent to the mechanism of hot pressing were also made. In many instances prismatic grains of silicon nitride, identified by electron diffraction, are enveloped by the yttrium-silicon oxynitride phase, for instance as in figure 1b. In exceptional cases such as figure 5a, the silicon nitride grains appear to be totally surrounded, and have formed with a highly prismatic morphology. The inter-facet angles are exactly 120°, with the facets being formed by the (10 $\bar{1}$ 0) planes of the silicon nitride. This is

vividly revealed by imaging one set of the  $(10\bar{1}0)$  planes as has been done in figure 5b. Here the facet plane is not only seen to be formed by a  $(10\bar{1}0)$  plane of atoms but also to be smooth without any steps at the scale of the interplanar spacing. Such pronounced faceting is generally characteristic of crystal growth from an isotropic liquid, indicative of a liquid phase densification process occurring during hot pressing. This conclusion is reinforced by the presence in the surrounding yttrium-silicon oxynitride grain of the low angle boundary indicated by the arrow (fig. 5a). This boundary extends from one corner of the silicon nitride grain in the micrograph all the way across the oxynitride grain until it terminates in a neighbouring silicon nitride grain. Such a boundary could only form as a result of crystallization around an already crystalline region (the silicon nitride grain).

Even in cases where the silicon nitride grains are not obviously surrounded as in the previous examples, they are still wet in part by the yttrium-silicon oxynitride. This however can only be revealed by stereo-microscopy techniques, and in particular by using the method known as 2 1/2 D dark field stereo-microscopy (16).

#### IV. Discussion

The observation of a thin interphase layer between the silicon nitride and yttrium-silicon oxynitride grains was unexpected because there is no evidence in the X-ray diffraction analyses of the material nor in previously reported analyses along the  $\text{Si}_3\text{N}_4 - \text{Y}_2\text{Si}[\text{Si}_2\text{O}_3\text{N}_4]$  tie line of a third phase. Therefore it is unfortunate that even with the techniques available it was not possible to

determine its composition. However, on the basis of the recently published behaviour diagram of Lange et al.<sup>(17)</sup> the phase in quasi-equilibrium with both silicon nitride and yttrium-silicon oxynitride could be  $\text{YSiO}_2\text{N}$ , recently determined to have the pseudo-wollastonite structure<sup>(18)</sup> or alternatively  $\text{Y}_4\text{Si}_2\text{O}_7\text{N}_2$  as reported by Wills<sup>(21)</sup>. The reason that the boundary phase could not be observed in the thicker regions of the sample is probably due to the difficulty in finding an interphase boundary that is both planar and oriented parallel to the electron beam throughout the thickness of the foil. In the very thinnest regions these conditions are naturally easier to satisfy.

The existence of a thin, possibly amorphous film, between the silicon nitride and the yttrium-silicon nitride grains suggests that its behaviour at high temperatures will profoundly influence the mechanical strength of the material as a whole. By analogy with MgO fluxed hot-pressed silicon nitride, the softening of the film at elevated temperatures would result in a loss in strength, and would contribute in part to the observed decrease in strength at high temperatures of the  $\text{Y}_2\text{O}_3$  fluxed silicon nitride.

The distribution of yttrium in the microstructure fully supports the findings of Rae et al.<sup>(7)</sup> that it is concentrated in the yttrium-silicon oxynitride phase. This work supplements that conclusion however by finding that no yttrium segregation in the silicon nitride occurs to within almost  $200\text{\AA}$  of the interphase boundary. When the shape of the different major phases and the observation that the impurity elements are all in solution in the oxynitride phase is considered it is clear that during hot-pressing a form of "solution-reprecipitation" mechanism had taken place. The silicon nitride crystallises out of solution,

rejecting impurity solute which is then trapped in solid solution during the subsequent cooling and solidification of the oxynitride phase. Such a mechanism, already proposed for densification in both the MgO hot pressed silicon nitride <sup>(19)</sup> and the  $\beta$ -sialon <sup>(20)</sup> systems, makes it possible to understand how the impurity concentration can be so appreciable in some grains and how it can vary so much within the same grain of yttrium-silicon oxynitride. Since the yttrium-silicon oxynitride is such a good sink for impurity elements, as suggested by Rae et al. <sup>(7)</sup> on the basis of its structural similarity with the melilite silicates, it is far from clear why the thin interphase phase should form at all. For this reason work is still in progress to identify the elemental composition of this phase.

## V. Conclusions

Three main conclusions concerning the microstructure of  $Y_2O_3$  hot pressed silicon nitride can be drawn from this work.

1. There is a third phase present in addition to the silicon nitride and the yttrium-silicon oxynitride. This is in the form of a thin (10-80 Å wide) film at the interphase boundaries and can be seen at all the boundaries in the very thinnest regions. It is speculated that the loss in high temperature strength is caused by this interphase phase in the same manner as the softening of the intergranular phase in MgO hot pressed silicon nitride is seen as the cause of its high temperature strength degradation.
2. All the yttrium, which was added in the form of  $Y_2O_3$ , is present in the yttrium-silicon oxynitride phase, with no yttrium segregation into the silicon nitride detectable within 180 Å of the interphase boundary.

8 8 8 8 4 7 6 1 5 6 3  
1 7 6 9

-11-

3. The yttrium-silicon oxynitride does dissolve appreciable concentrations of impurity elements in the starting powders, notably Ti, W, Fe, Ni, Co, Zn, in addition to the Ca, Mg and Al found by Rae et al. This fact, the location of the yttrium and the morphology of the yttrium-silicon oxynitride and silicon nitride grains suggests that densification occurs by a "solution-precipitation" mechanism as has been found in the MgO hot pressed system (19).

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FIGURE CAPTIONS

Fig. 1. The yttrium-silicon oxynitride phase (the darker phase) is generally observed at multiple silicon nitride grain junctions, as arrowed in Fig. 1a. It is also to be seen partially enveloping prismatic shaped grains of silicon nitride, P, as in Fig. 1b. Bright field transmission electron micrograph, 100kV.

Fig. 2. Microanalysis trace A, showing only elements Y and Si, was obtained from the yttrium-silicon oxynitride phases as illustrated in Fig. 1a. (The Cu peaks arise from the copper grid on which the sample was mounted). Trace B was also obtained from an oxynitride phase but from one morphologically similar to that of Fig. 1b rather than Fig. 1a. The central part of trace B is expanded and labelled in the lower diagram.

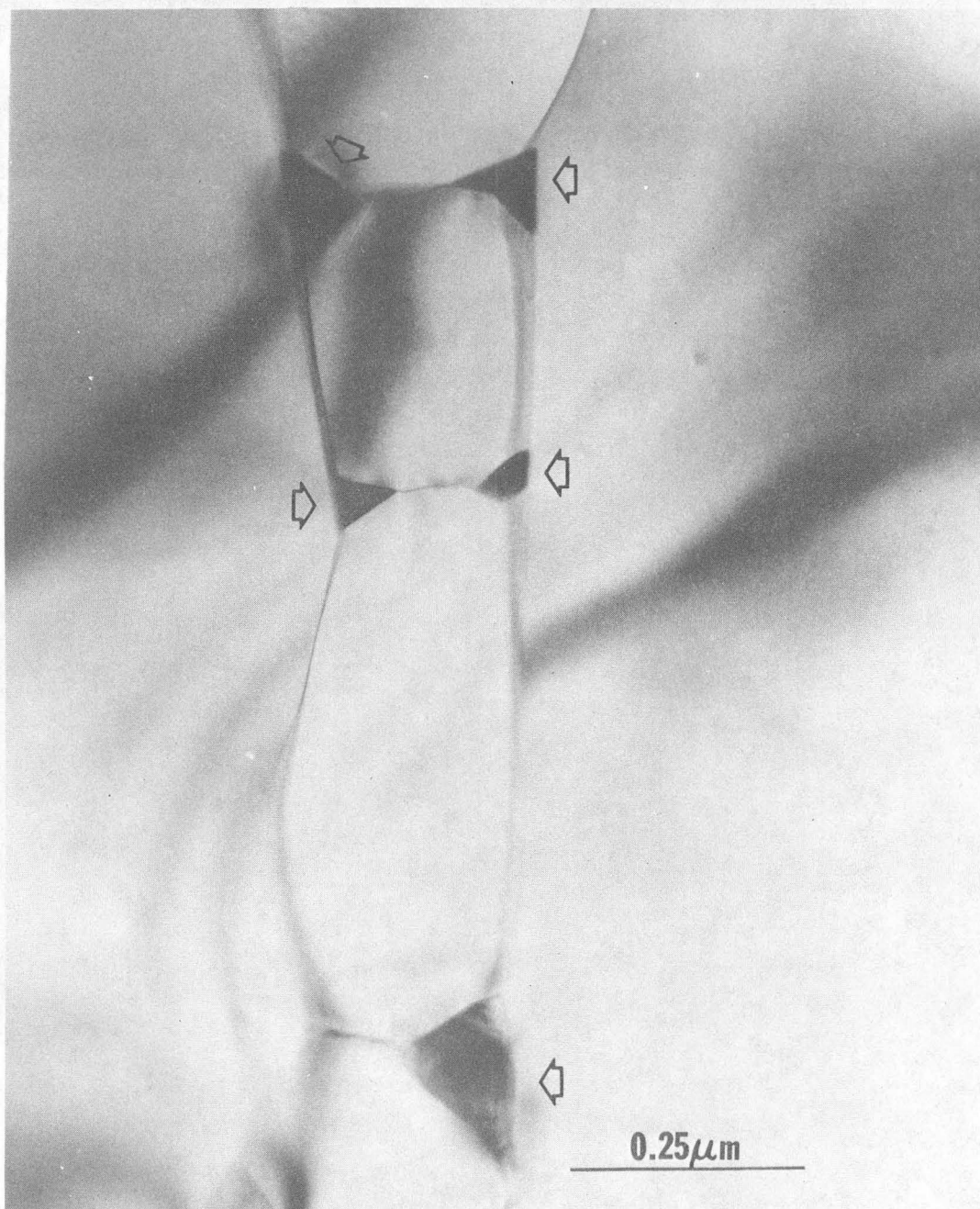
Fig. 3. A thin intergranular phase between adjacent grains of silicon nitride and yttrium-silicon oxynitride. The phase, appearing as thin white lines and arrowed, can only be observed in the thinnest regions. Bright field transmission electron micrograph, 100kV.

Fig. 4. The discontinuity of lattice fringes at the interphase boundary between a beta silicon nitride grain (left) and a yttrium-silicon oxynitride grain (right) confirms the presence of a third phase, which at point A is  $26\text{\AA}$  wide. A lattice fringe image transmission electron micrograph, 100 kV.

Fig. 5a. A relatively perfect prismatic grain, P, of beta silicon nitride surrounded by a matrix of yttrium-silicon oxynitride. The shape of the grain and the presence of the sub-grain boundary (arrowed) leading from the silicon nitride grain at the bottom left suggests

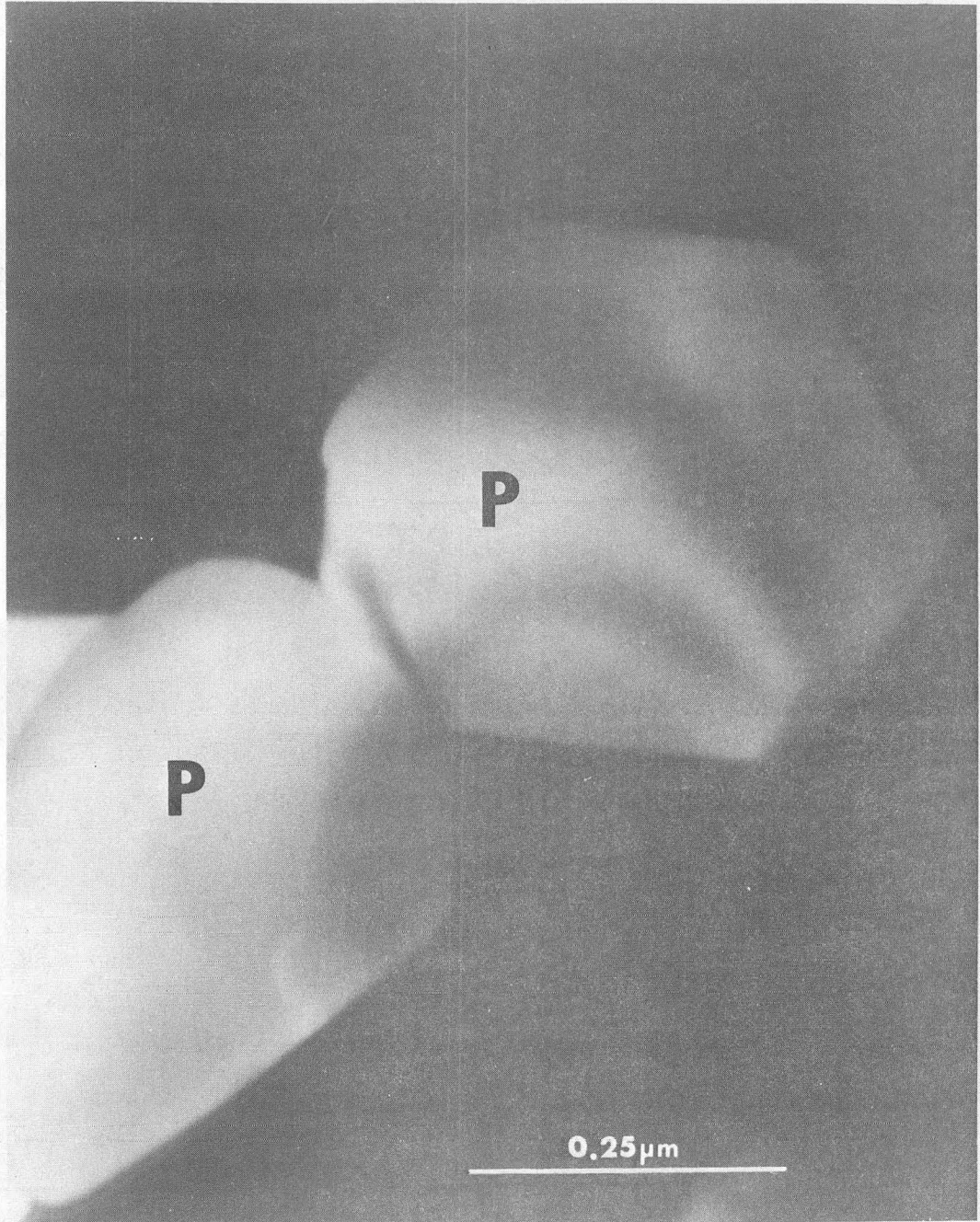
that the silicon nitride formed by a "solution-reprecipitation" mechanism. Lattice fringe image electron micrograph, 100kV.

Fig. 5b. Lattice fringe image of part of the interface between the silicon nitride grain, P, of Fig. 5a, and the yttrium-silicon oxynitride, S. The facet is seen to be remarkably smooth and to be bounded by a  $(10\bar{1}0)$  plane of the silicon nitride. 100kV.



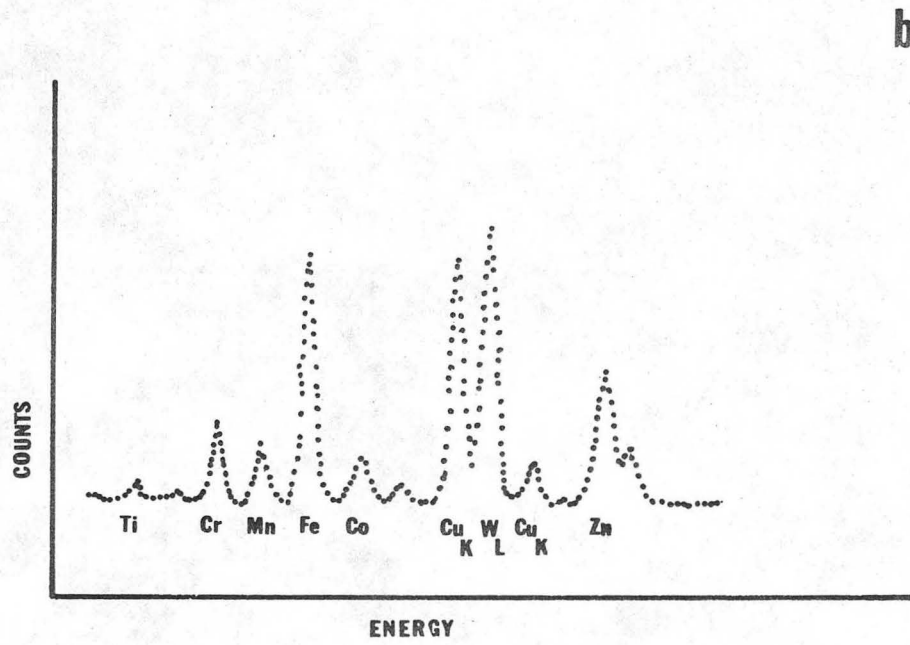
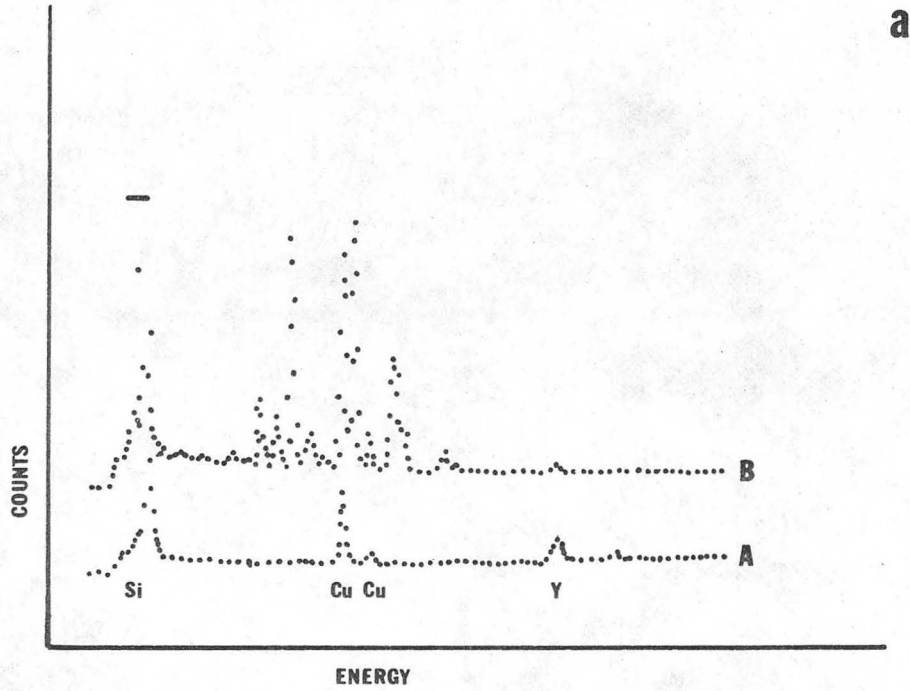
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FIG. 1a



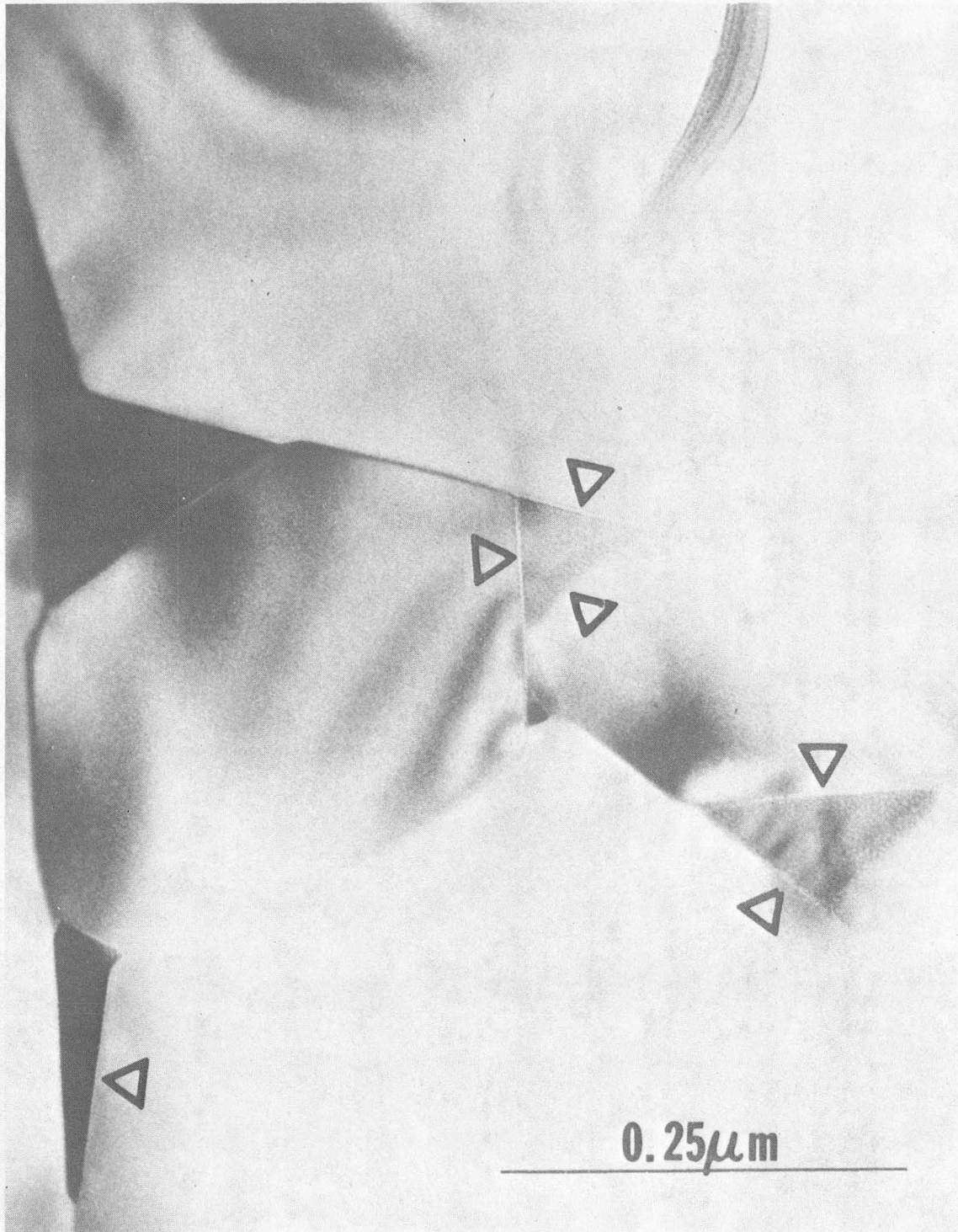
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FIG. 1b



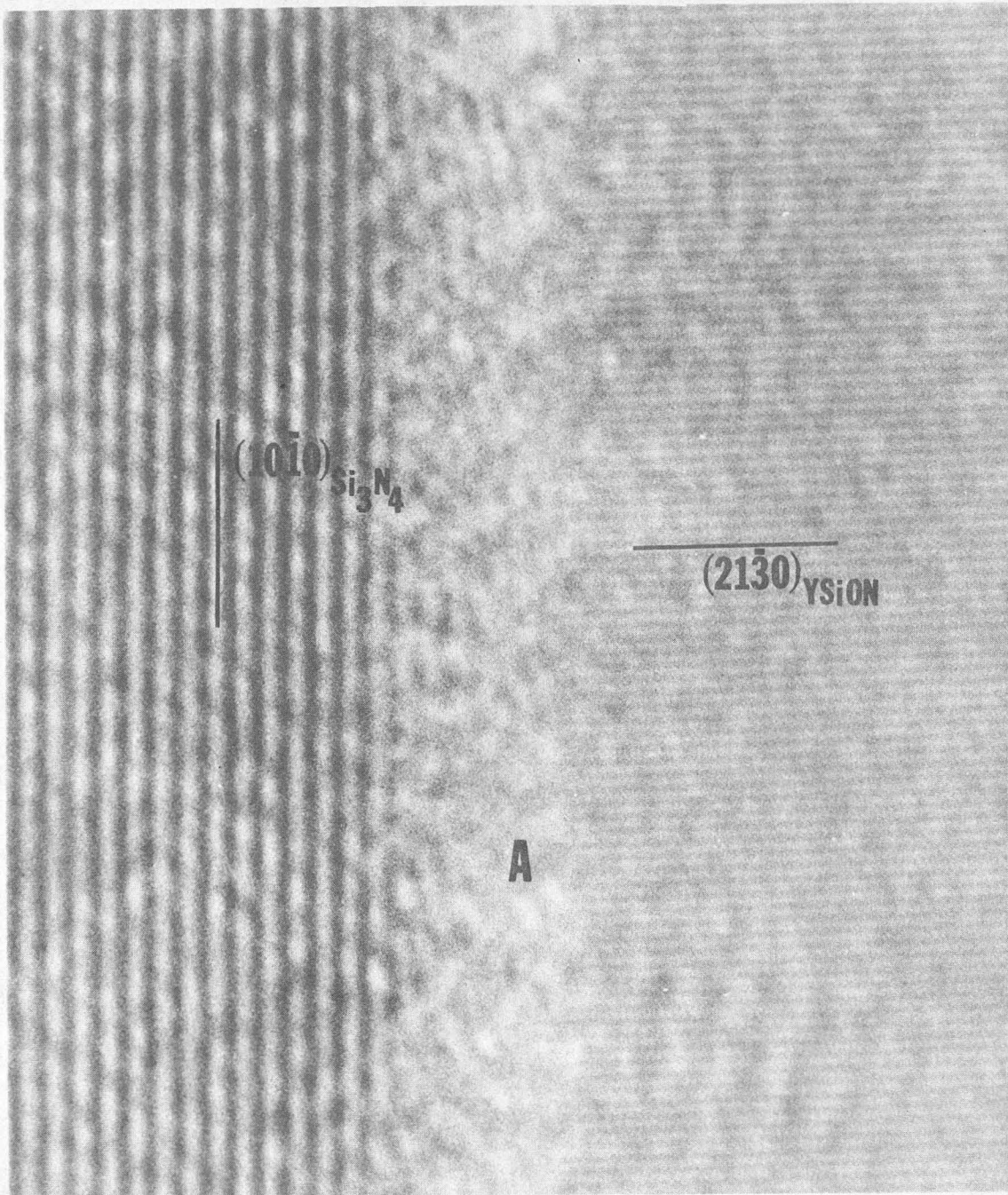
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FIG. 2



XBB 776-5857

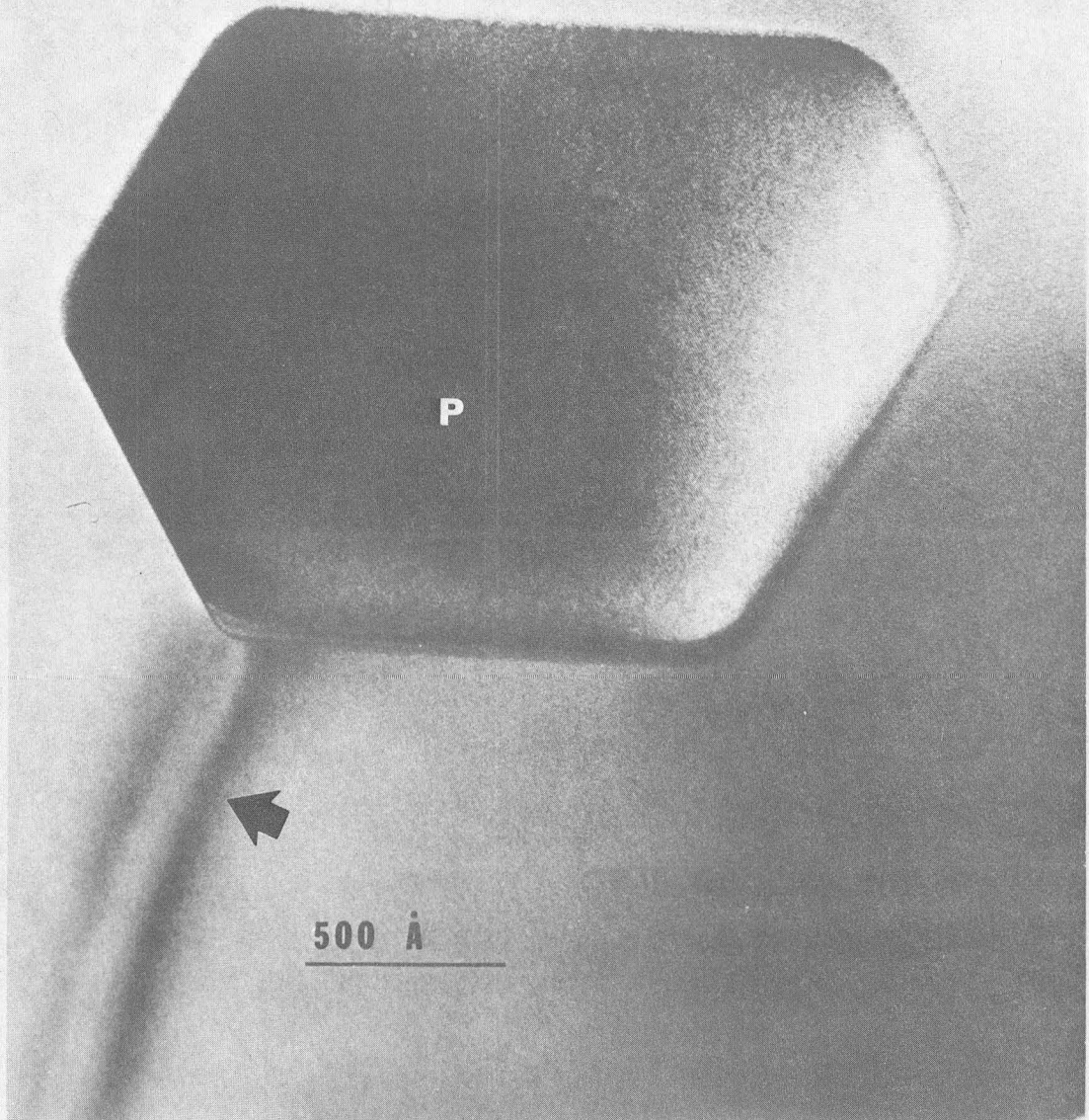
FIG. 3



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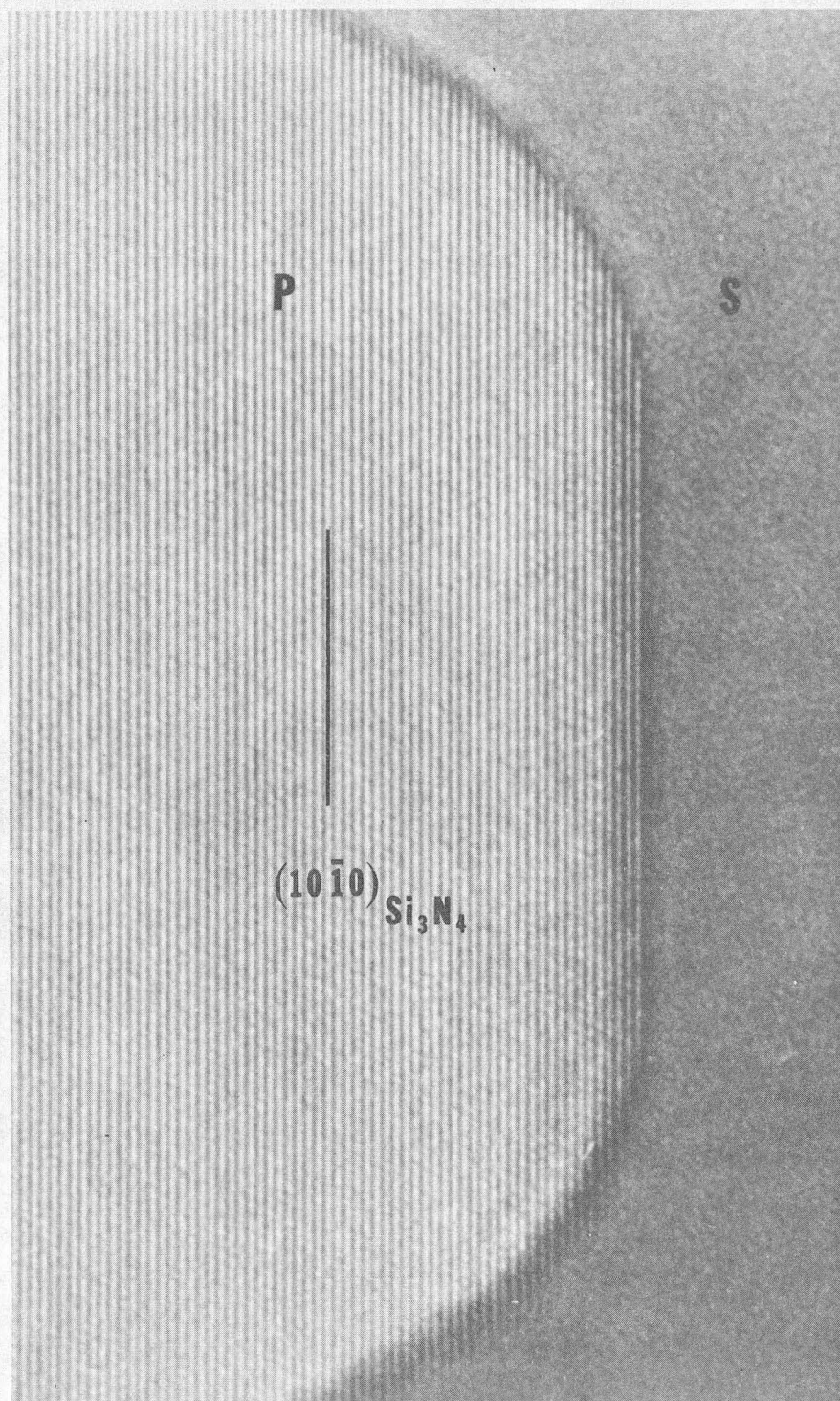
FIG. 4





XBB 772-779

FIG. 5a



XBB 772-783

FIG. 5b

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