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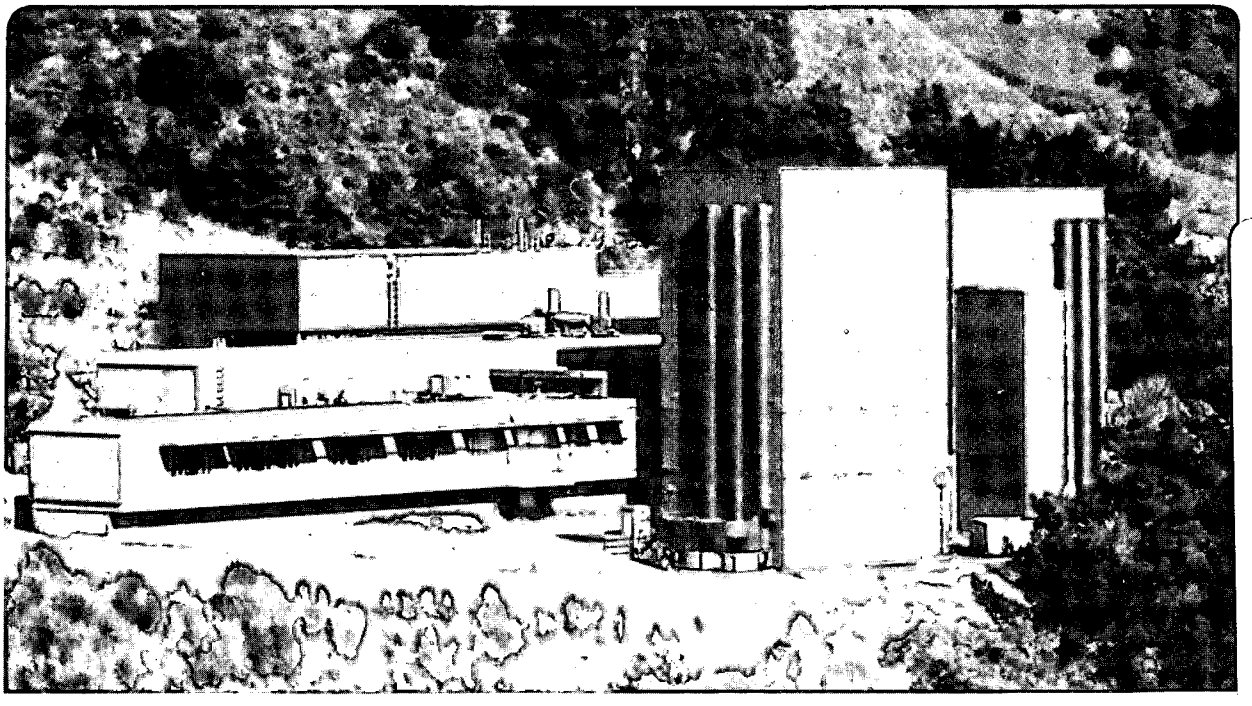

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Epitaxial Growth of (011) Al on (100) Si by Vapor Deposition

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Abstract

The morphology, orientation relationship and interface structure of Al vapor-deposited on (100) Si single crystal substrates were investigated by X-ray diffraction and transmission electron microscopy. It was shown that vapor growth at room temperature results in a random (111) texture whereas growth at 280°C leads to films with high-quality (011) epitaxy and a high degree of grain boundary faceting. Due to alignment of close-packed directions in the plane of the interface there are two orientation variants with a morphology characterized by an oriented 90° <011> mazed bicrystal structure.

Heteroepitaxial growth of Al on Si single crystal substrates depends on the orientation of the substrate and the conditions of growth [1-6]. For (100) substrates high-quality films have been grown by the ionized cluster beam technique [7]. Their microstructure has been characterized by transmission electron microscopy [8,9] as that of a mazed bicrystal thin film with many grains in only two orientations separated by a network of grain boundaries devoid of triple junctions. Later it was shown that under ultrahigh vacuum (UHV) conditions, films of comparable quality and with the same texture could be produced by room temperature molecular beam epitaxy (MBE) growth on (100) substrates with 1x2 surface reconstruction [10]. More recently it was reported that even sputter deposition of Al-1%Si at 200°C in a vacuum of 7×10^{-7} mbar leads to the same epitaxy found previously [11,12]. In this letter we show that films of better quality can be obtained by simple physical vapor deposition at 280°C, whereas room temperature deposition leads to films with a random (111) texture. The films were characterized experimentally by X-ray diffraction (XRD) and transmission electron microscopy (TEM).

Al films of thickness 100-300 nm were vacuum deposited from an Al charge of 99.999% purity on single crystal p-type Si (100) wafers under a base pressure of 2×10^{-7} mbar. The substrates were cleaned by initially oxidizing in a solution of 1:1 by volume hydrogen peroxide and sulfuric acid for about 10 minutes, cleaning with deionized water and finally etching for about 10 min in a solution of 10% HF in deionized water, followed by a water rinse. Prior to and during deposition, the substrates were heated to 280°C. At about 100 nm/s the deposition rate was extremely high. The texture of as-deposited films was characterized by XRD and the epitaxial alignment, the in-plane orientation and the film quality were examined by TEM in plan view and cross section. Plan view TEM samples were prepared by cutting 3mm discs, mechanical dimpling from the substrate side to less than 10 μm and Ar ion thinning to electron transparency. Cross section samples were made using the method described by Bravman and Sinclair [13].

Fig. 1a shows an XRD scan typical for a film deposited at room temperature, exhibiting a strong (111) texture evidenced by the absence of all but the (111) peak of Al. At increased substrate temperature a pronounced (011) texture appeared. This is illustrated in figure 1b which shows an XRD scan after extended (4h) isothermal holding of the substrate at 280°C prior to

deposition. It is evident from this diffraction scan that the $(111)_{Al}$ peak is now absent and has been replaced by a strong $(022)_{Al}$ peak.

In figure 2, the corresponding electron diffraction patterns show the in-plane alignment of the films. Films deposited at room temperature give rise to polycrystalline rings formed by $(220)_{Al}$ reflections superimposed on the square spot pattern of the (100) Si substrate (see figure 2a). As expected for a (111) textured material $(111)_{Al}$ and $(200)_{Al}$ reflections are absent. Hence this film has a (111) texture with random in-plane arrangement. Figure 2b shows the in-plane alignment of films deposited at 280°C. In addition to the square substrate (100) pattern there are two $(011)_{Al}$ patterns rotated by 90° relative to each other. The orientation relationship is one in which close packed $<01\bar{1}>$ directions in the substrate and overlayer are parallel in the interface, while the (011) planes of Al are parallel to the (100) surface of the Si substrate, i.e.:

$$(011)_{Al} \parallel (100)_{Si} \qquad \text{and} \qquad [01\bar{1}]_{Al} \parallel [01\bar{1}]_{Si}.$$

This relationship is identical to that reported previously for different growth modes. There are two orientation variants of the (011) Al pattern because there are two equivalent ways of orienting (011) Al on (100) Si in such a way that the close packed directions are aligned. This is illustrated schematically in figure 3a. Figure 3b shows a diffraction pattern of a thin film after the Si substrate has been removed. For ease of interpretation one of the patterns is marked with black dots. The 90° misorientation between the two orientation variants (black and white) is clearly visible. Also shown in this image are the traces of mirror symmetry planes of the combined color symmetry pattern [14]. It is apparent that vertical and horizontal mirror planes exchange the black and white variants while the diagonal mirrors reflect each variant into itself. The point symmetry of this microstructure is characterized by the $\underline{4}m\underline{m}$ tetragonal magnetic point group. Here an underlined symmetry element denotes a color exchange operation [15].

In figure 4 this structure is shown in a complementary pair of bright field images, taken under mirror-related imaging conditions in which only one orientation variant at a time is in strong diffracting condition and thus appears dark. The two orientation variants with random distribution form the "Holstein"-type grain structure characteristic for these mazed bicrystal films without triple

grain boundary junctions. It is apparent from this figure that extensive faceting has taken place. As observed for films deposited by the ionized cluster beam technique the preferred facets lie on the mirror planes of the substrate [9]. The degree and topology of this faceting is a simple and direct measure of the anisotropy of the grain boundary structure [14].

Fig. 5 shows an image obtained in an orientation nearly perpendicular to the substrate. A similar spacing between the (220) planes in the Si substrate and the (200) planes in the Al film leads to a moiré pattern seen as an array of parallel lines. Because the mismatch between these sets of parallel planes is about 5% the moiré pattern magnifies all misorientations and distortions about 20 times [16]. The moiré pattern is thus a good measure of the degree of perfection of these films. Using this effect, the axial misorientation between grains was found to be in the range of 90±1°, a better in-plane epitaxial alignment than that of 90±2° reported for sputter-deposited films [10,11]. The azimuthal misorientation was measured from microdiffraction patterns taken across individual grain boundaries. It was found that the $[011]_{Al}$ plane normals deviated by no more than 1° from each other.

The Al/Si interface was examined directly in cross-sections prepared from these films. Figure 6a shows a view of a cross section from a film about 200 nm thick. It is interesting to note that the grain boundaries are all very nearly perpendicular to the substrate.The average spacing between grain boundaries is about 100 nm. An almost periodic contrast is visible at the substrate/film interface. Close examination revealed that this is not due to misfit dislocations but is most likely related to the presence of amorphous pockets or islands. This is more clearly visible (regions marked by arrows) in the high resolution micrograph shown in figure 6b. The two crystals are viewed along their common close-packed direction. The Si substrate is in zone axis orientation whereas the Al film is seen to be slightly off its [011] zone axis, due to a slight axial misorientation. In addition, the Al lattice is misoriented slightly with respect to the Si lattice by a rotation of about 1° around the incident beam direction (azimuthal misorientation). This can be seen by viewing the image at an oblique angle. Because the amorphous pockets remained visible through an entire through-focal series (of which figure 6b shows only one member) and because

similar pockets were observed in many independent locations at intervals corresponding to the near-periodic contrast seen in figure 6a, it was concluded that these amorphous islands are part of the interface structure obtained with the present procedure. It is likely that epitaxial growth is initiated through pinholes in the oxide layer on the substrate surface. The remaining oxide islands get buried during film growth.

In summary, we have shown that physical vapor deposition of Al at 280°C leads to the same (011) epitaxy on (100) Si, but with better epitaxial alignment and with a much higher growth rate than that achieved by other, technically more involved techniques.

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Figure Captions

Fig. 1 XRD scans comparing Al film texture on (100) Si substrates during vapor deposition at room temperature (a) and at 280°C (b) showing a clear change from a strong (111) texture to a pronounced (011) texture.

Fig. 2 Electron diffraction patterns corresponding to figure 1, illustrating the in-plane (axial) alignment, changing from random orientation about a common <111> axis (a) to a well-defined epitaxial bicrystal orientation (b).

Fig. 3 Schematic illustration of the two variants marked L and R of the observed orientation relationship (a) and bicrystal diffraction pattern after substrate removal (b). One orientation variant in the bicrystal pattern is marked with black dots.

Fig. 4 Complementary pair of bright field images illustrating the "Holstein" type microstructure, its faceted grain boundaries and its symmetry.

Fig. 5 Bright field image with moiré fringes illustrating degree of perfection of orientation and faceting.

Fig. 6 Cross section view of 200 nm thick film showing planar interface and regular arrangement of grain boundaries perpendicular to the interface (a); and high resolution image showing presence of amorphous pockets and confirming slight lattice misalignment (b).

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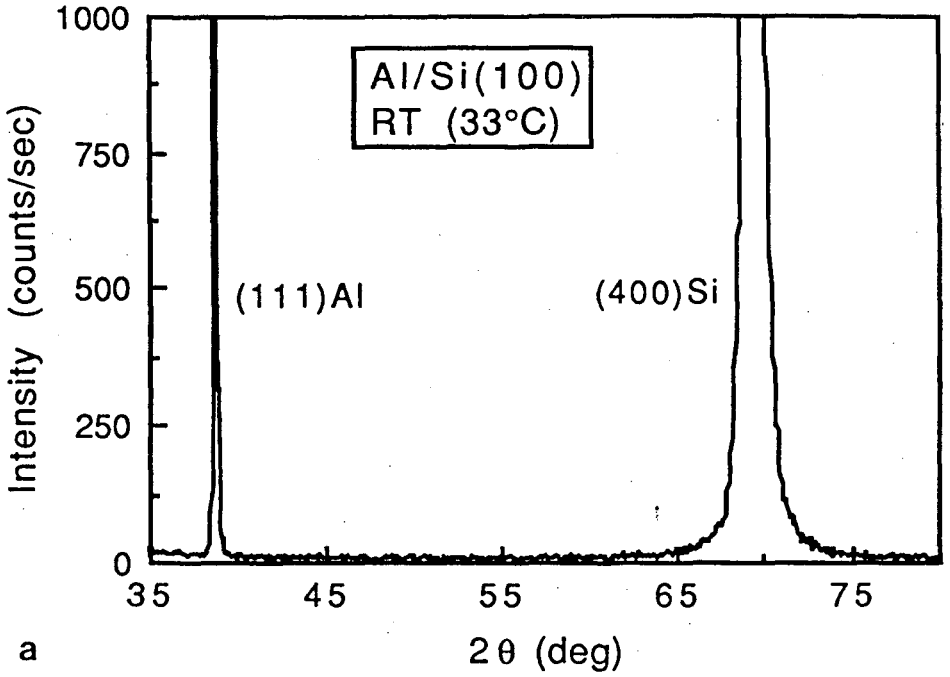

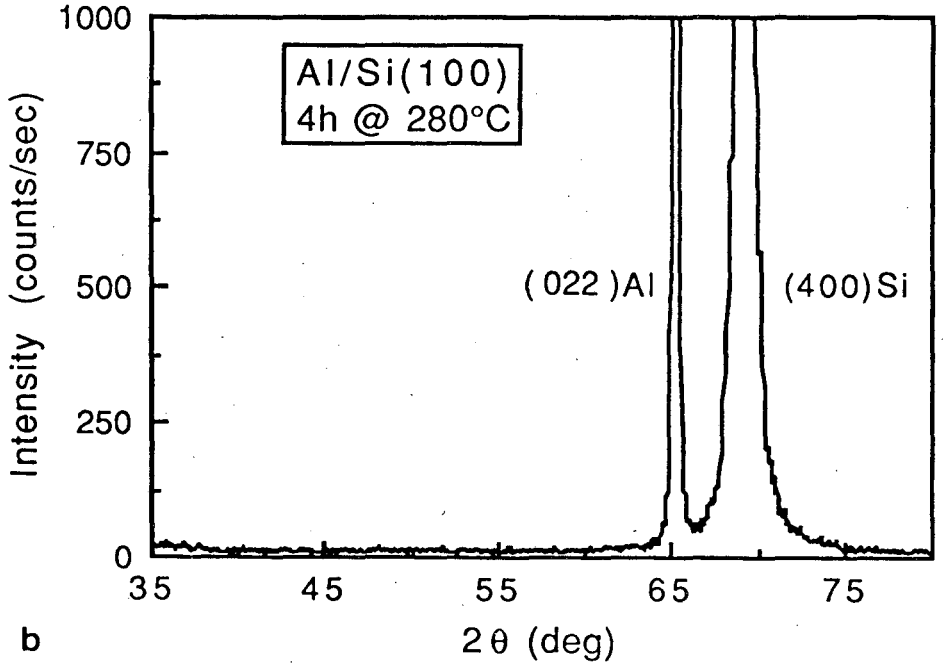

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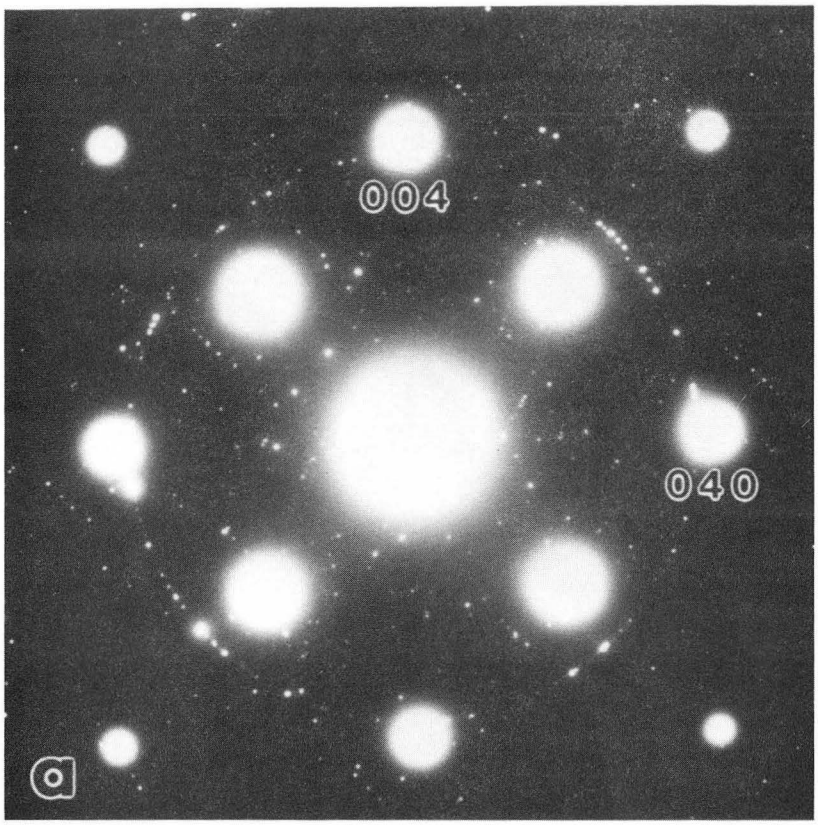
Figure 1



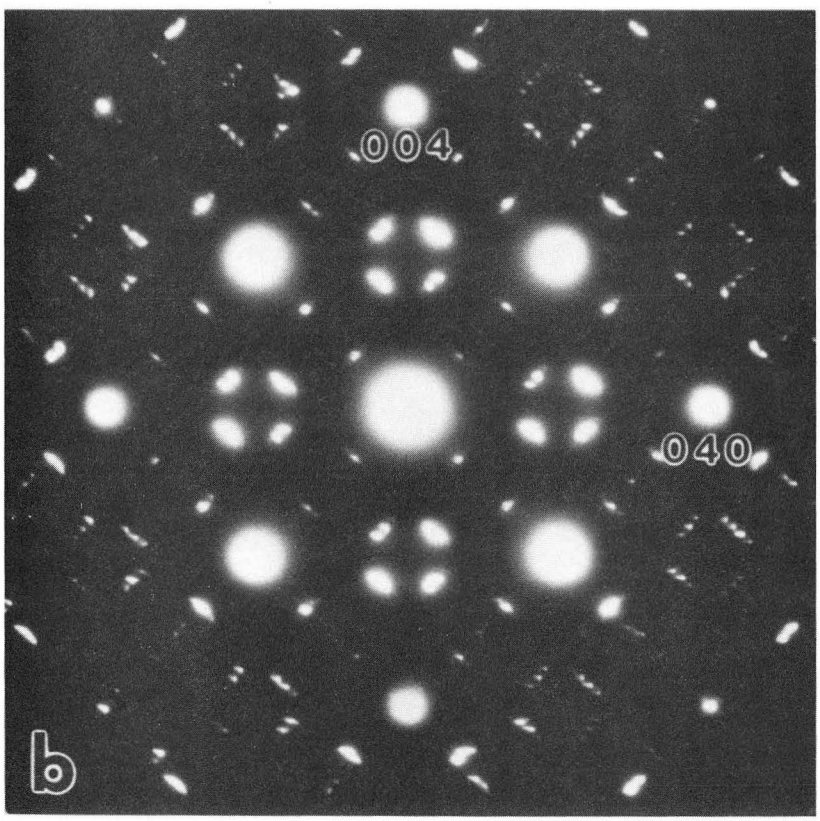


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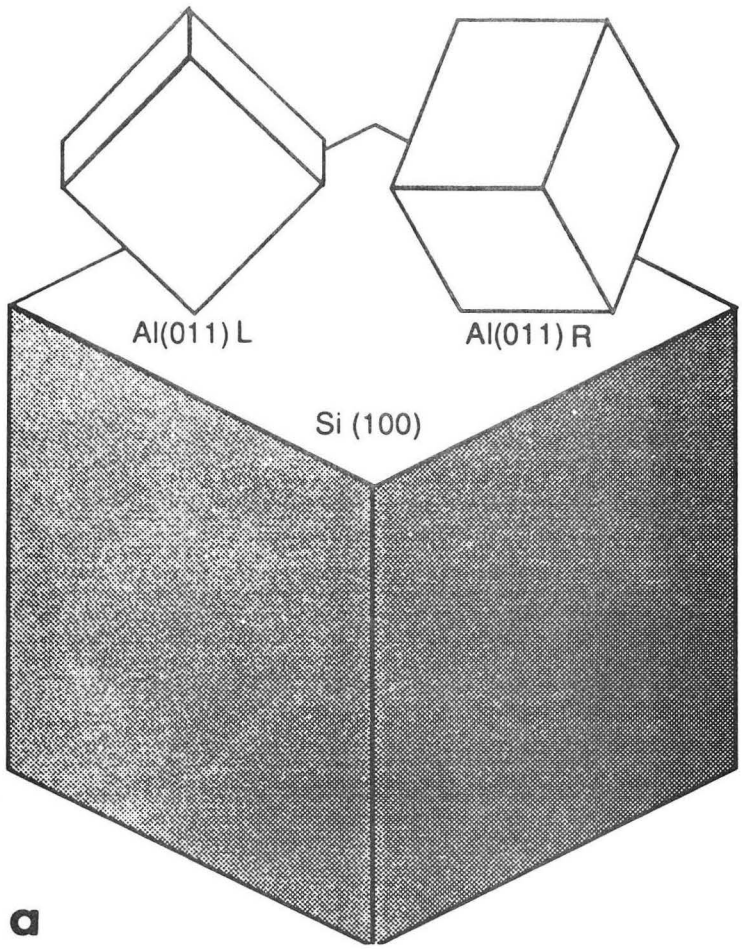
Figure 2



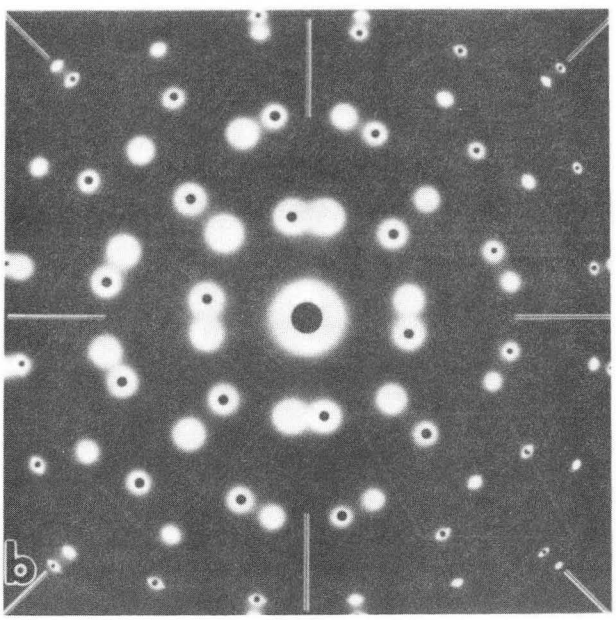


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Figure 3

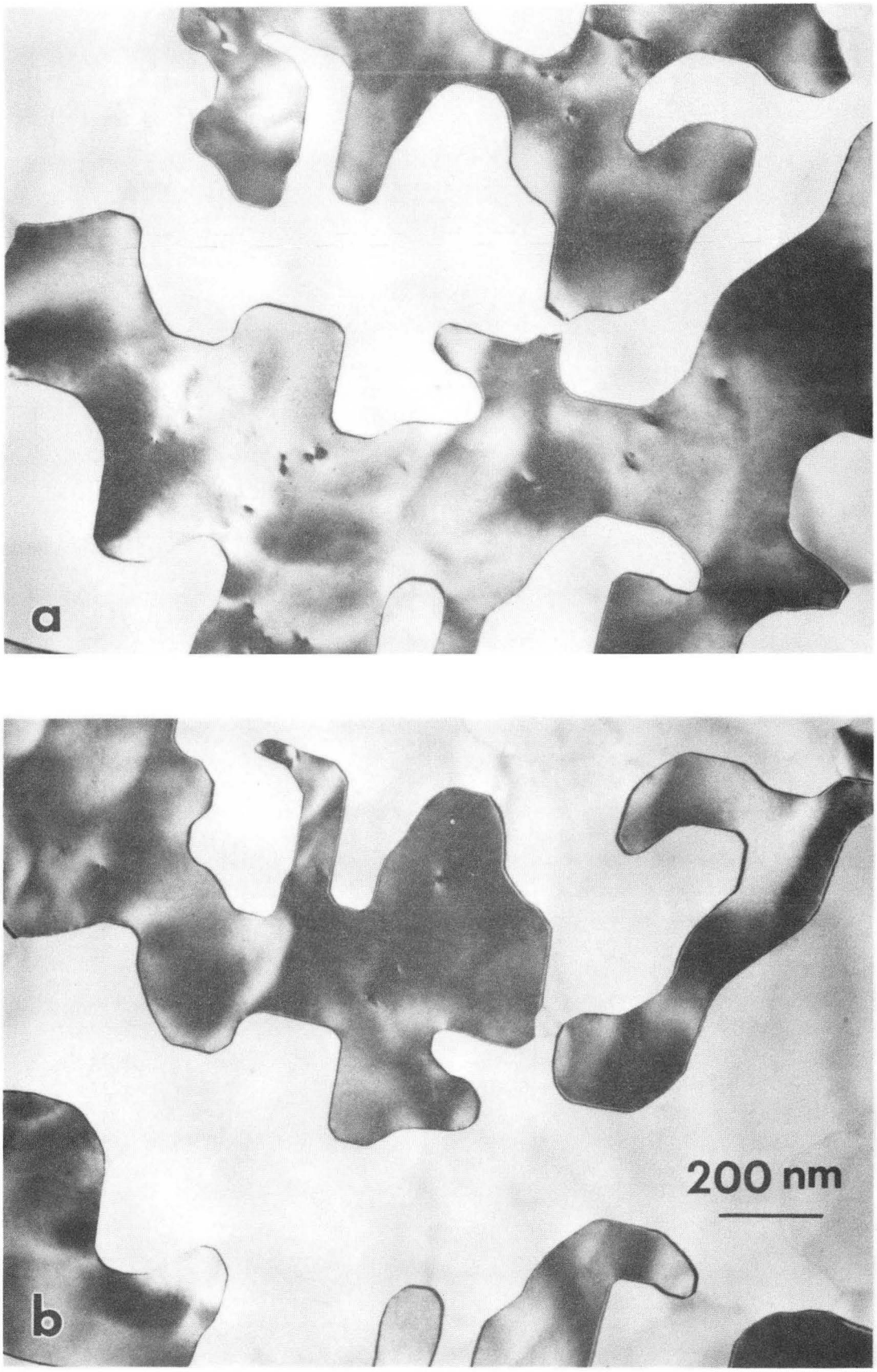

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Figure 4

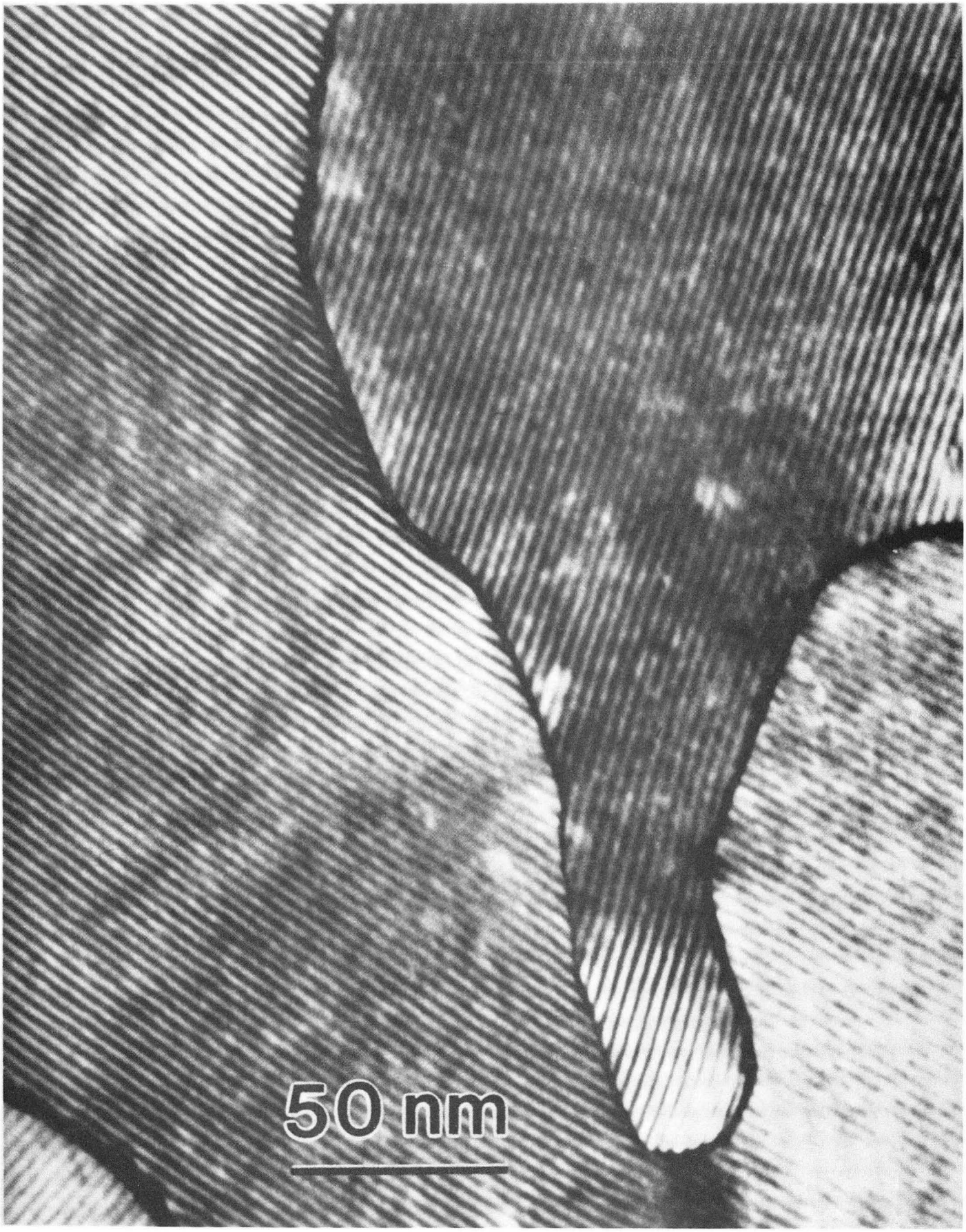

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Figure 5

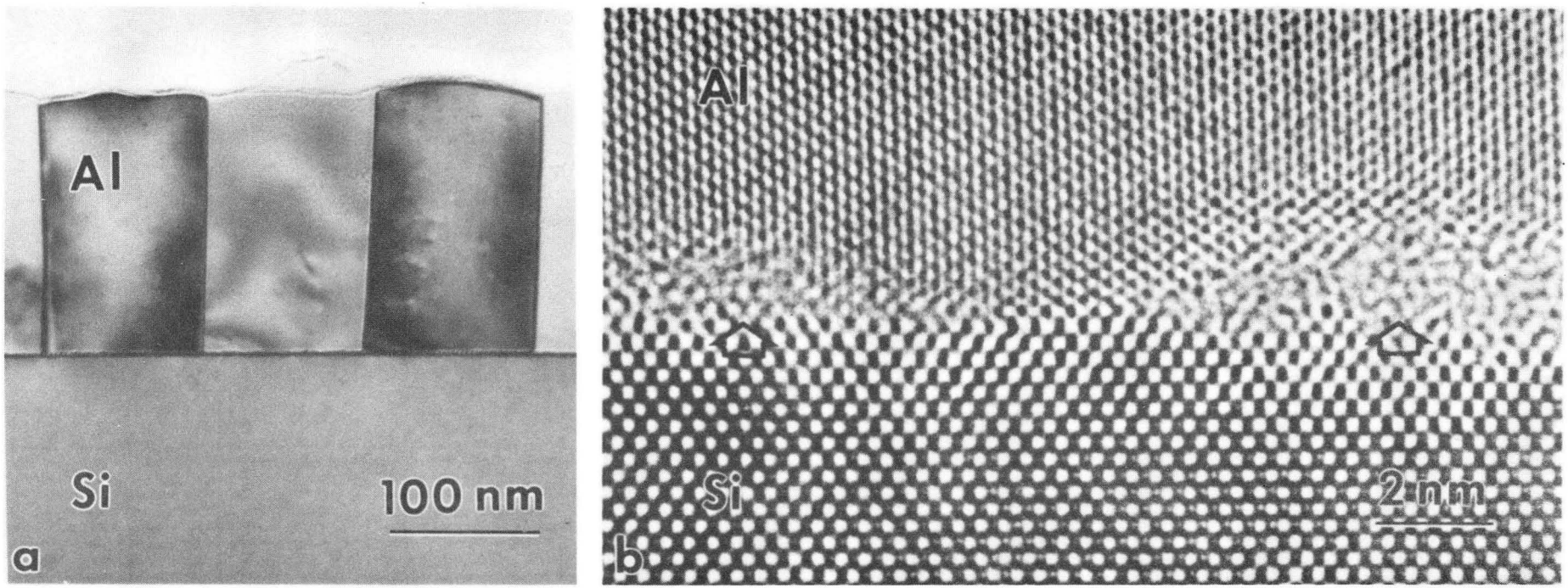

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Figure 6

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