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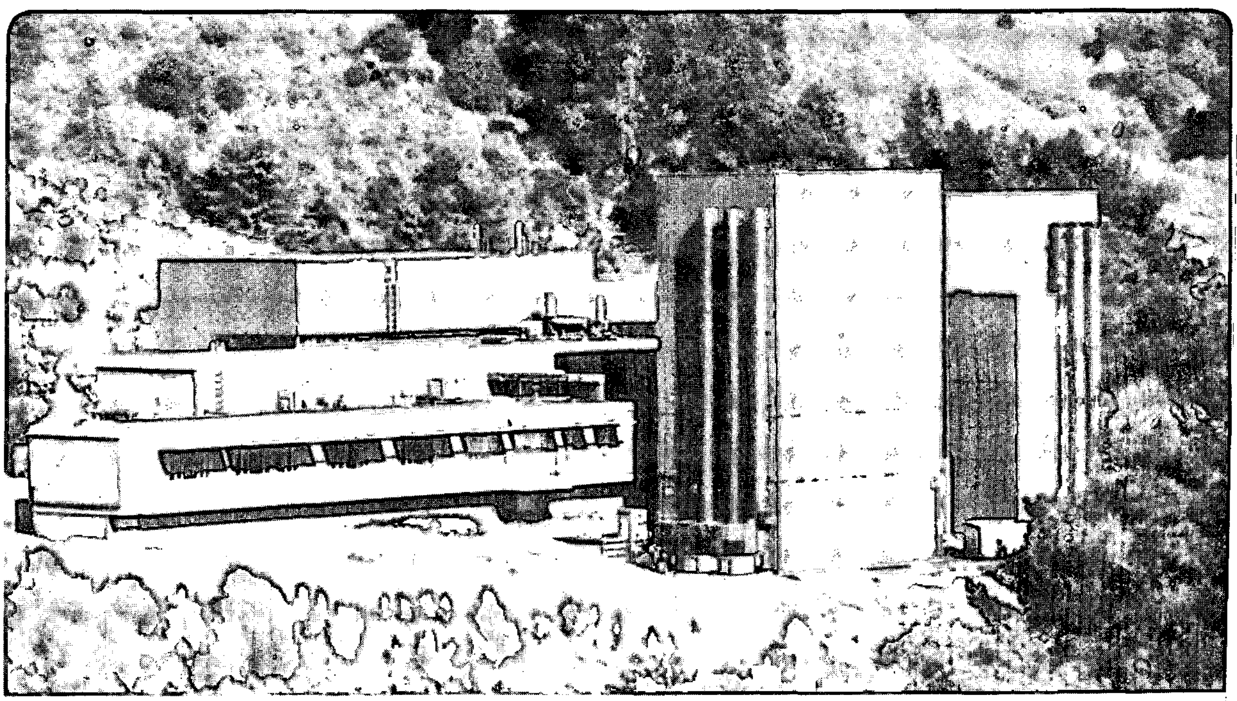

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TWIN FORMATION IN Ag SEEDED Co/Pt MULTILAYERS GROWN ON GaAs BY MBE

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ABSTRACT

Molecular beam epitaxy (MBE) was used to grow ultrathin Co-Pt multilayers on GaAs (111) substrates with 200Å thick Ag layer as a buffer. Magnetic properties (B-H loop) of the multilayers, measured by a vibrating sample magnetometer (VSM), confirmed that these samples exhibit strong anisotropy perpendicular to the film surface. Twin-related Ag grains were found to nucleate on the substrates by reflection high energy electron diffraction and low-energy electron diffraction. Epitaxial relationship of the multilayers with respect to the substrate was investigated by (high-resolution) transmission electron microscopy . Twin-related grains, 30-40 nm in diameter, are present in the multilayers. These twins are generated either by propagation of the twin boundaries in the Ag layer into the multilayers or by nucleation of twin-related Pt grains on the Ag buffer surface.

INTRODUCTION

There has been a growing interest in multilayers of magnetic and non-magnetic materials for the past decade due to their unusual magnetic properties and potential device applications. These properties include perpendicular magnetic anisotropy [1-3], giant magnetoresistance [4], antiferromagnetic coupling [5], and long range oscillatory coupling [6]. The strong perpendicular magnetic anisotropy in multilayers of a few atomic layers of Co or Fe and Pt or Pd, persists to high temperatures and has technological applications in magneto-optical information storage [7].

Perpendicular magnetic anisotropy of these films has generally been attributed to the reduced symmetry [8] at the interfaces between magnetic and non-magnetic materials as well as limited interdiffusion [9] or combined with partial alloy ordering [10,11] at the interfaces. In recent studies, structural defects such as stacking-faults [10] and planar defects along the growth direction [12] have been discussed as possible sources for the strong magnetic anisotropy. The exact origin, however, remains unclear. In particular, Co/Pt multilayers grown on GaAs substrates by MBE exhibit significantly different magnetic properties depending on growth orientations such as (111), (110), and (100). This suggests that simple arguments of symmetry breaking at the interfaces are an inadequate description and a more critical evaluation of the microstructure is required.

To understand the origin of the perpendicular magnetic anisotropy exhibited by such multilayers, we have been investigating the crystallography and microstructure of these structures. In this report, we discuss the origin and growth of twin related grains in Co/Pt multilayers grown on (111) GaAs with a 200 Å thick Ag buffer layer. The initial growth of the Ag seed film was investigated in situ by RHEED and low energy electron diffraction (LEED). The magnetic properties of the multilayers were measured by a VSM. The microstructure of the multilayers was studied by conventional TEM using both plan and cross-section view samples. The crystallography of the twins and their propagation, as well as the epitaxial relationship between the various components of the multilayer stack, were investigated by HRTEM.

EXPERIMENTAL

A VG 80-M MBE system was used to prepare Co/Pt multilayers on GaAs substrates. The substrates were cut with the surface normal within ~0.5^{o} of the [111] direction. The specific face used in these experiments was the [111] B-face, i.e., the As-terminated face. The substrate surface was polished mechanically and chemically. The substrates were then heated to ~600^{o}C to remove surface impurities and to generate a 1x1 RHEED pattern. The Co and Pt layers were grown from e-gun sources at rates of 0.15 and 0.25 Å/sec, respectively. The background pressure before and during the film growth were approximately $2x10^{-11}$ mbar and $2x10^{-10}$ mbar, respectively.

Multilayer stacks of 3Å Co and 15 Å Pt, with either 15 or 30 repeats, were grown on the GaAs substrates. A 200 Å thick Ag film was first deposited onto the GaAs surface held at 100^{o} C. This film grew with its [111] axis parallel to the GaAs [111] axis and served both to seed the subsequent growth of Pt along [111] and to prevent chemical reactions between Pt and the GaAs substrate. The multilayer stack was grown onto the Ag beginning with Pt. A Pt capping film was deposited onto the final Co film to help prevent oxidation of the structure. Additionally, a similar 75 Å sputtered film of Si_3N_4 provided further protection. Figure 1a shows a schematic diagram of the multilayer.

RHEED and LEED patterns were recorded during the film growth. The electron energy for RHEED was 12.6 keV and for LEED was varied from 19 to 400 eV. In the RHEED measurements the incidence angle was in the range 1-3 degree.

Conventional TEM study of the multilayers was carried out using a JEOL 200CX TEM operating at 200 kV. Plan-view specimens were prepared from 3 mm diameter discs cut from the MBE grown samples. These discs were polished from the GaAs-side until the thickness was 100 μm. After grinding and polishing, the backside was dimpled to a thickness of ~15 μm, and then ion-milled with 5 kV Ar ions using a liquid nitrogen-cooled specimen stage. Cross-section specimens were prepared by cutting the samples into rectangular strips, 2 mm in width, gluing two strips with the deposited films facing one another, and slicing the glued strips into 3 mm lengths. These were thinned and polished to a thickness of approximately 100 μm from both sides perpendicular to the interface between the substrate and the film. The thinned samples were dimpled and ion-milled in the same way used for the preparation of the plan-view specimens. High-resolution images were recorded using the Atomic Resolution Microscope (ARM) at the National Center for Electron Microscopy, operating at 800 kV. High-resolution images were obtained over a wide range of defocus values of the objective lens and with the electron beam parallel to the [110] orientation of the specimens. The spherical aberration (C_S), and point to point resolution of the microscope was 1.4 mm and 0.17 nm, respectively.

B-H loops were measured on both the 15 and 30 repeat Co/Pt multilayers at room temperature using a VSM. The applied magnetic field strength ranged from -10 kOe to +10 kOe. The sensitivity of the induced magnetization was 10^{-3} emu. The sample size was ~ 5 mm x 5 mm.

RESULTS

The hysteresis loop from the 30 period sample (figure 1b) exhibits a squareness ratio (Bm/Br: Bm is maximum magnetic induction, Br; remanence) of 0.97. Coercivity and retentivity of the multilayers are 3726 emu and 1.1×10^3 Oer, respectively.

RHEED patterns recorded during the nucleation and growth of the initial Ag film are shown in figure 2. The GaAs surface, after heat-cleaning, exhibited 1x1 symmetry with diffraction streaks elongated along direction of the rods. Surface roughness is indicated by the streaks, partially separated along the rod direction. Additionally, along azimuths +60 degree, -60 degree, and 180 degree rotated from the [112] azimuth the specular beam was bifurcated. This indicates the development of (110) facets possibly as a result of thermal etching. Corresponding LEED patterns confirmed the 3-fold symmetry of the surface and the presence of facets. At 0.1 monolayer (ML) coverage of Ag, streaks with the spacing of bulk Ag are evident. These increase in intensity up to 4 ML where the GaAs streaks are present but barely visible. This suggests that the Ag nucleates as islands and that complete coverage of the GaAs is attained between 4 and 10 ML. The fact that all the Ag RHEED patterns reproduced on a 60 degree rotation of the sample rather than 120 degree indicates rotational twinning of islands about the [111] pole. This finding was confirmed by LEED which showed 6-fold symmetry of the spot pattern at all energies unlike bulk single crystals of Ag which showed 3-fold symmetry.

A bright field image and the corresponding selected area diffraction pattern recorded from a plan-view specimen are shown in figure 3. The Co/Pt multilayers is polycrystalline and consists of 30-40 nm diameter grains. An amorphous phase, ~1 nm wide, is present at the grain boundaries. However, the SAD pattern, in figure 3b, shows a six fold symmetry, with each spot consisting of reflections with a deviation less than 5°.

Figure 4 shows one of the high-resolution images recorded from a cross-section of the 15 period multilayer sample. This image was obtained from an area of a thickness less than 100 Å, near the Scherzer defocus value of -550 Å. The lattice spacings of the Ag and the Co/Pt layers were examined by optical diffractrogram; these spacings were matched to those of perfect Ag and Pt crystals with an error of ~ 1 % . The {111} planes of the Ag, and the Co/Pt layers are parallel to {111} GaAs. On the other hand, {111} planes of grains M and N in the Co/Pt layers indicate that one grain is rotated with respect to the other by 70.5° about the [110] axis. An edge-on type boundary adjoining grains M and N is seen along the line indicated with arrows u and v. Moire fringes resulting from the superposition of twin related grains are seen in both the Ag and the Co/Pt layers. A {112} twin boundary indicated with an arrow w in the Ag layer appears to propagate through the Co/Pt multilayers. Local lattice distortion is seen at the interface between the substrate and the Ag layer. Contrast from the periodicity of ~18Å, corresponding to the multilayer period, clearly observed in X-ray measurements of both low angle reflectivity and high angle diffraction [13] in the Co/Pt layers is not observed.

DISCUSSION

The RHEED and LEED data confirmed that Ag nucleates as islands on GaAs (111) with two distinct orientations related by twinning. The twins correspond to two different stacking sequences along the [111] axis: ABCABC... or BACBAC..., respectively and are related by a 180 degree rotation about this axis. The similar intensity of the 6-fold spots in the LEED patterns indicates a roughly 50:50 distribution of the two orientations. This distribution is maintained throughout the Ag film growth.

The presence of twin related grains in the multilayer stack was independently confirmed by transmission electron microscopy. An optical diffractrogram obtained from the area indicated with a rectangle in figure 4 is shown in figure 5. A periodicity of 6.7 Å along the (111) direction is seen. This periodicity arises due to double diffraction from the twin related grains. Twinned grains along two crystallographic directions are generated for (111) growth of face centered cubic films. Their overlap in the cross-section view may either lead to Moire fringes in the image, or alternatively, to the observed 0.149 $Å^{-1}$ spatial frequency in the optical diffractograms, figure 5a.

Similar Moire fringes were observed in samples of Co/Pt multilayers by Chien et al [10]. However, they interpreted this contrast as arising from stacking faults generated in an $L1_2$ phase of equiatomic composition formed by atomic mixing at the interface. A model based on such faults was also presented to suggest a mechanism for the origin of uniaxial magnetic anisotropy. Their microstructural interpretations and defect-related model are inconsistent with our interpretation of the contrast in the high resolution and plan view micrographs.

In plan view, the bright field image is polycrystalline while the SAD pattern corresponds to a single crystal structure. This contradiction can be resolved in our interpretation. The two twin grains are generated by 60^o rotation about the (111) axis of one grain with respect to the other. Therefore, the diffraction pattern recorded with the [111] orientation of the sample will remain unchanged and the six fold symmetry is preserved.

The films exhibit strong [111] texture. A low angle deviation less than 5^o about the [111] axis and $< 2^o$ along the [111] axis is observed in the corresponding diffraction patterns. The latter is believed to be very small, considering the parallel alignment of the (111) plane of the Co/Pt layer to the (111) GaAs plane in the cross-section high-resolution transmission electron micrographs. The amorphous phase at the boundaries may be due to phase segregation or oxidation along the boundaries. Further work is underway to determine the chemistry of the phase at the boundaries, as this may play a critical role in the modelling of the magnetic anisotropy of these films.

It has been reported [12] that Co/Pt multilayers grown on GaAs substrates by magnetron sputtering also show a columnar texture consisting of planar defects perpendicular to the surface, and these films exhibit good perpendicular anisotropy. The samples investigated in our study show higher squareness, compared to the sputtered samples. It is interesting to see the presence of a columnar texture both in our samples and the sputtered multilayers, even though the MBE sample has a much smaller angular deviation along the surface normal, from one grain to the other, than the sputtered multilayers. Further investigations, such as growth of twin-free multilayer stacks, either by growth on single crystal Pt substrates or on GaAs wafers off-cut from the exact (111) direction by a few degrees, and a more detailed characterization of the twin morphology, is required to understand the origin of perpendicular anisotropy in these multilayers.

CONCLUSION

The Co/Pt multilayers grown on GaAs (111) substrates with a 200 Å thick Ag layer as a buffer by MBE exhibit a strong magnetic anisotropy. The initial growth stage of the Ag buffer, and the overall microstructure, in particular, twin formation in the Co/Pt multilayers, were investigated by RHEED and TEM, respectively. The Ag buffer layer nucleate in an island morphology on GaAs substrate at 100°C and a twin-relation is found to exist among the nuclei. Twin domains, 30-40 nm in diameter, are also present in the overall Co/Pt multilayer stack. Coherent twin boundaries parallel to the (111) substrate were observed. On the other hand, boundaries 1 nm in width propagate through the multilayers. These boundaries contain an amorphous phase and exhibit little preferential crystallographic orientations. Interpretation of the origin of perpendicular anisotropy in such seeded multilayers should incorporate these microstructural features in any proposed models.

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FIGURE CAPTIONS

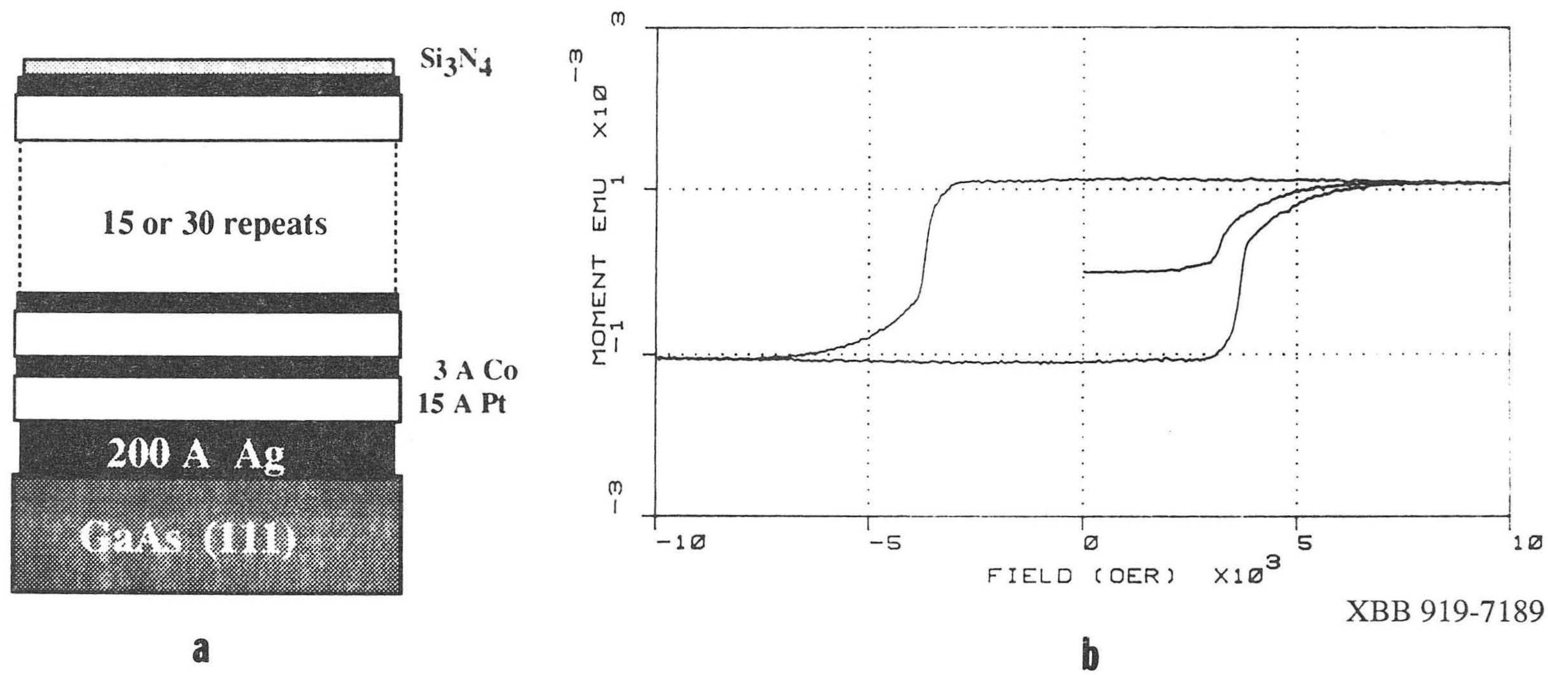
Figure 1. (a) Schematic representation of the Co/Pt multilayer stack, including the Ag buffer layer, grown on GaAs (111) substrates. The films terminated with a Pt layer and were capped with 75 Å of Si_3N_4 to prevent subsequent oxidation. (b) B-H loop of the 30 repeat Co/Pt multilayers.

Figure 2. In-situ RHEED patterns during the growth of Ag in (a) [111], (b) [112] directions.

Figure 3. (a) Bright field image of a plan-view specimen. (b) the corresponding selected area diffraction pattern.

Figure 4. High resolution image of a cross-section view specimen. Twin associated Moire fringes are seen in both the seeded Ag and Co/Pt multilayers. A {112} twin boundary in the Ag layer propagates into the multilayer stack along the line indicated with an arrow 'w'.

Figure 5. (a) Optical diffractogram obtained from the area indicated with a rectangle in figure 4. (b) Schematic of the diffractogram in figure 5a.



Si3N4
15 or 30 repeats
3 A Co
15 A Pt
200 A Ag
GaAs (111)
MOMENT EMU X10^-3
3
1
-1
-3
-10
-5
0
5
10
FIELD (OER) X10^3
XBB 919-7189
a
b

Figure 1

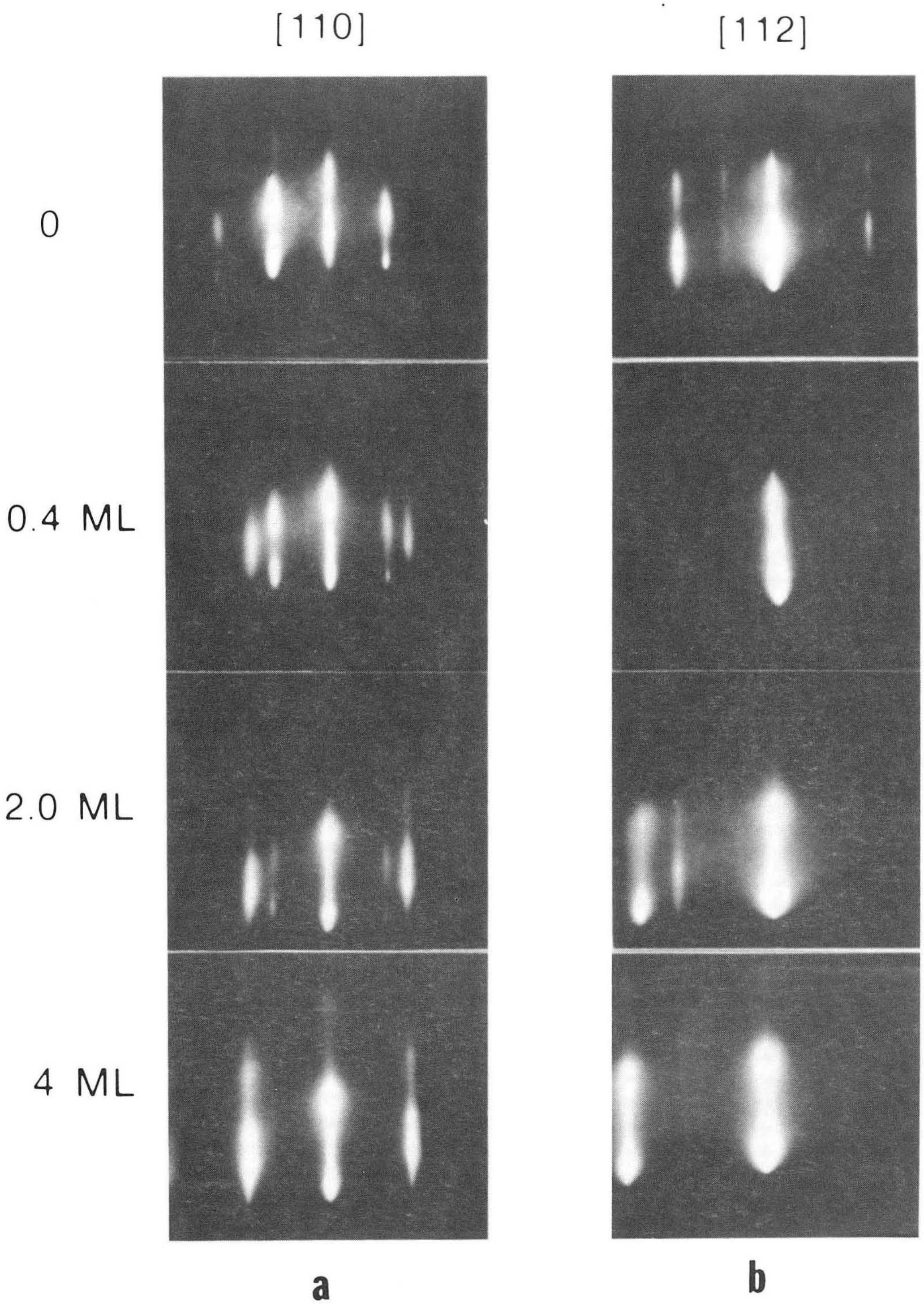


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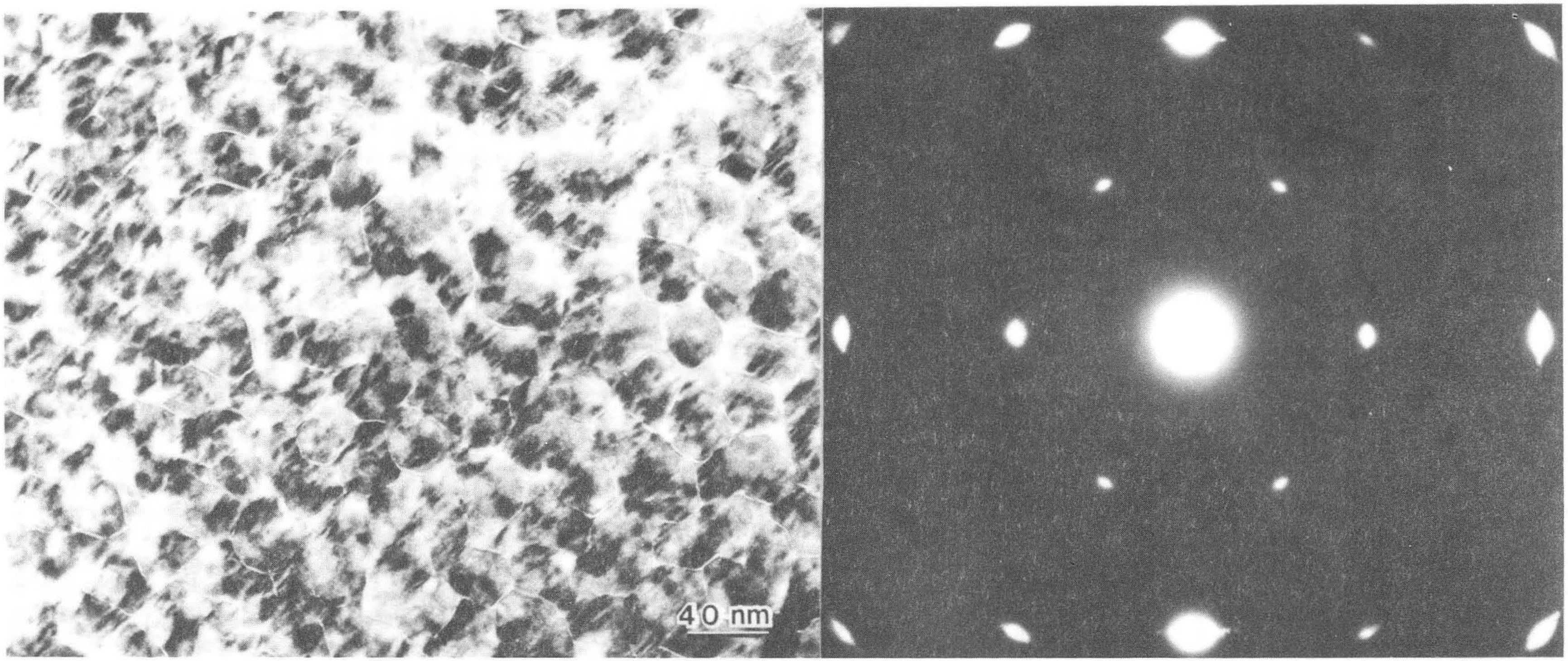
Figure 2



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a b

Figure 3

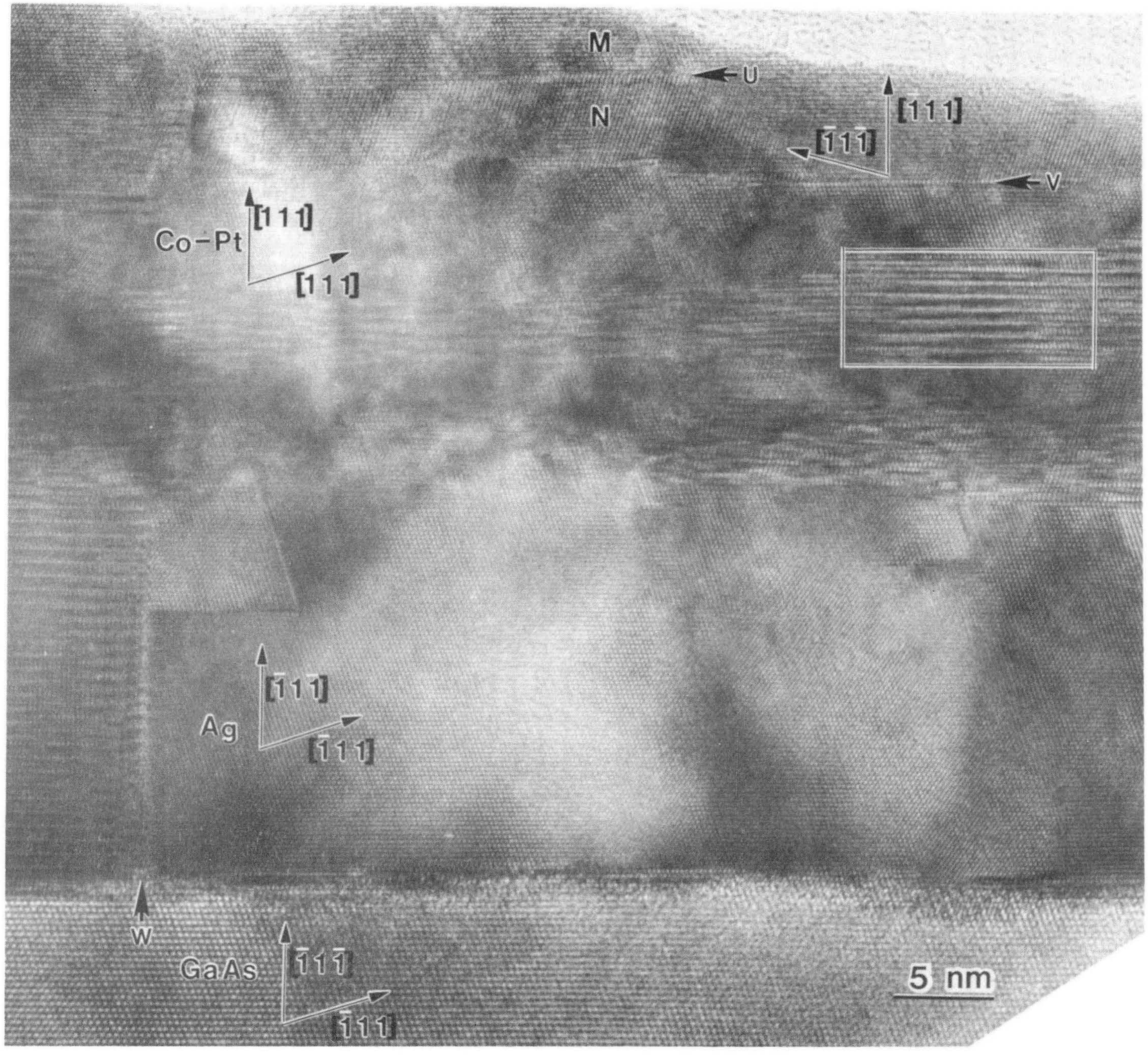

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Figure 4

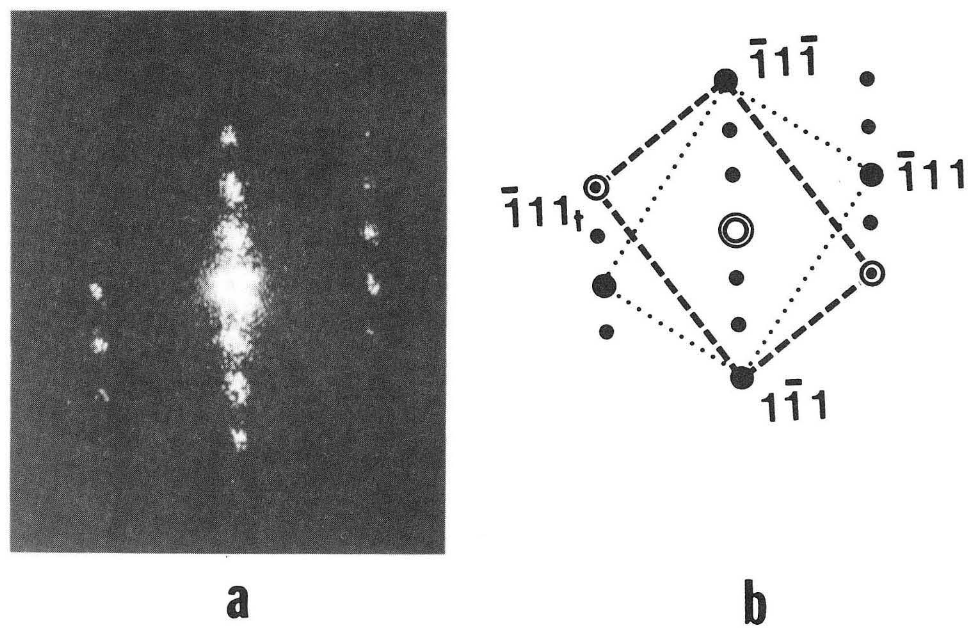

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Figure 5