Pergamon

Acta mater. Vol. 46, No. 7, pp. 2299–2303, 1998

PII: S1359-6454(97)00385-6
1359-6454/98 $19.00 + 0.00

BUFFER LAYER MORPHOLOGY EFFECTS ON THE ORDERING OF EPITAXIAL FePd(001) THIN FILMS

P. CARO, A. CEBOLLADA, D. RAVELOSONA, J. TAMAYO, R. GARCíA and F. BRIONES

IMM (CNM/CSIC) and UAM, Isaac Newton 8 PTM, 28760 Tres Cantos, 28006 Madrid, Spain

Abstract—The structural properties of thin FePd(001) films grown on MgO(100) substrates by UHV triode-sputtering have been studied as a function of the morphology of the substrate/seed layer interface. Changes in short range ordering are found to be correlated with the thickness of the Pt seed layer which controls the interface morphology and the formation of order domains in the FePd alloy along the growth direction. © *1998 Acta Metallurgica Inc.*

1. INTRODUCTION

New nanostructured magnetic materials with extraordinary properties such as giant magnetoresistance, oscillatory magnetic coupling, or large perpendicular magnetic anisotropies can be synthesized in single crystalline thin film form by means of epitaxial techniques such as molecular beam epitaxy (MBE). Surface kinetics aspects related to the particular growth technique and growth conditions used are very important for determining structural and consequent physical properties of the nanostructures. Obviously, an important factor for epitaxy is the compatibility of substrate lattice parameter and crystal symmetry with that of the grown layer. However, many recently published results show also the determinant role of the specific properties of the substrate/epitaxial layer interface. In particular, in order to grow crystalline metallic magnetic films on insulating substrates of technological impact like semiconductors or oxides, an appropriate conditioning or buffering of the interface is necessary.

Several substrates are used for growing metallic thin films, like oxides, such as MgO, zirconia or sapphire. Many of these materials have typically big terraces on which a seed layer of a metal is grown at a certain temperature, to obtain a flat, single crystalline material. For example, Lairson *et al.* [1] have shown that single crystalline Pt thin films with different orientations can be grown by planar magnetron sputtering onto heated substrates. They have found that under their growth conditions, a 0.3 nm Fe layer grown onto MgO prior to deposition of Pt yields to (001) epitaxial films with no (111) orientations. On the other hand, McIntyre *et al.* [2, 3] find that e-beam evaporated Pt films have a mixture of crystallites having (111) and (001) orientation over a range of substrate temperatures from 25 to 700°C, with the (111) orientation dominant at low substrate temperatures. This epitaxial growth is also found in MBE Pt (001) films grown at 700°C on MgO(100) [4]. It is obvious that the strength of the interfacial chemical bond is important in the kind of growth of a metal on a ionic crystal, and it has been compared for the Au/MgO *vs* Au/NaCl system [5].

Once the seeding epitaxy technique for metals is controlled, the study of many materials systems with different crystalline orientations and structures can be performed. For the magnetic materials, and both from the basic research and technological points of view, research in systems exhibiting both high magnetocrystalline anisotropy and magneto-optic effects is of great importance. Chemical ordering is a relevant topic in the study of metallic binary alloys in which one of the constituents is magnetic. New magnetic properties, such as strong magnetic anisotropies or enhancement in the magneto-optical activity, are predicted [6] and observed in the presence of an ordered phase [7–13].

Ordering in polycrystalline or single crystalline bulk samples is usually obtained by prolonged annealing of disordered alloys, making use of bulk atomic diffusion. Examples of alternative procedures are the annealing of multilayered samples [8] or the atomically controlled MBE deposition of alternating species [9–11]. In the same way as in the fabrication of semiconductor ordered alloys, codeposition of the elements at elevated substrate temperatures can lead to highly ordered, single crystalline metallic alloys [7, 12, 13]. Direct deposition on MgO does not lead to an ordered phase [13]. The interface has to be modified by adding a Pt seed layer to allow the correct epitaxy.

In our particular case, FePd alloys are known to adopt the ordered CuAu (I) structure. It consists of monoatomic planes of pure Fe and Pd stacked along the (001) direction in a face centered cubic lattice, in contrast to the disordered phase that is a solution of randomly positioned Fe and Pd atoms

on a fcc lattice. We have observed that cosputtered FePd(001) ordered alloys can be obtained in the CuAu (I) structure and show perpendicular magnetic anisotropy as a consequence of this ordering [13]. We will show here how the morphology of the buffer layer, and therefore its interface with the substrate and the film, strongly determines the structure of the alloy grown on top.

2. EXPERIMENT

The deposition of the films was performed in a UHV triode-sputtering system with a load-lock entry. The base pressure was in the low 10^{-7} Pa range and the Ar pressure during deposition was 4×10^{-2} Pa. Deposition rates were around 0.01–0.03 nm/s, depending on the target. The structure of the films was checked *in situ* with Reflection High Energy Electron Diffraction (RHEED) and *ex situ* with X-ray diffraction (XRD) using Cu Kα radiation. Atomic Force Microscopy (AFM) has been used to characterize the morphology of the Pt seed layers in air. The nominal composition was determined by careful calibration of the targets sputtering rates and checked by electron micro-probe, confirming the equiatomic ratio within an error of ~5%. The resulting thicknesses were cross checked using a profilometer.

The samples were grown on polished MgO(100) single crystals purchased from Marketech International, with a maximum misorientation of 0.5° and a RMS roughness of 0.4 nm. Substrates were cleaned in successive ultrasonic baths of trichloro-ethane, acetone and methanol. After being transferred to UHV, the MgO substrates were annealed at 700°C for several hours using a resistive heater. Pt seed layers of various thickness (5, 15, 30 and 45 nm) are first grown at 700°C. Following, a 100 nm thick FePd film is grown on top of the Pt at 500°C by codeposition of Fe and Pd from inde-

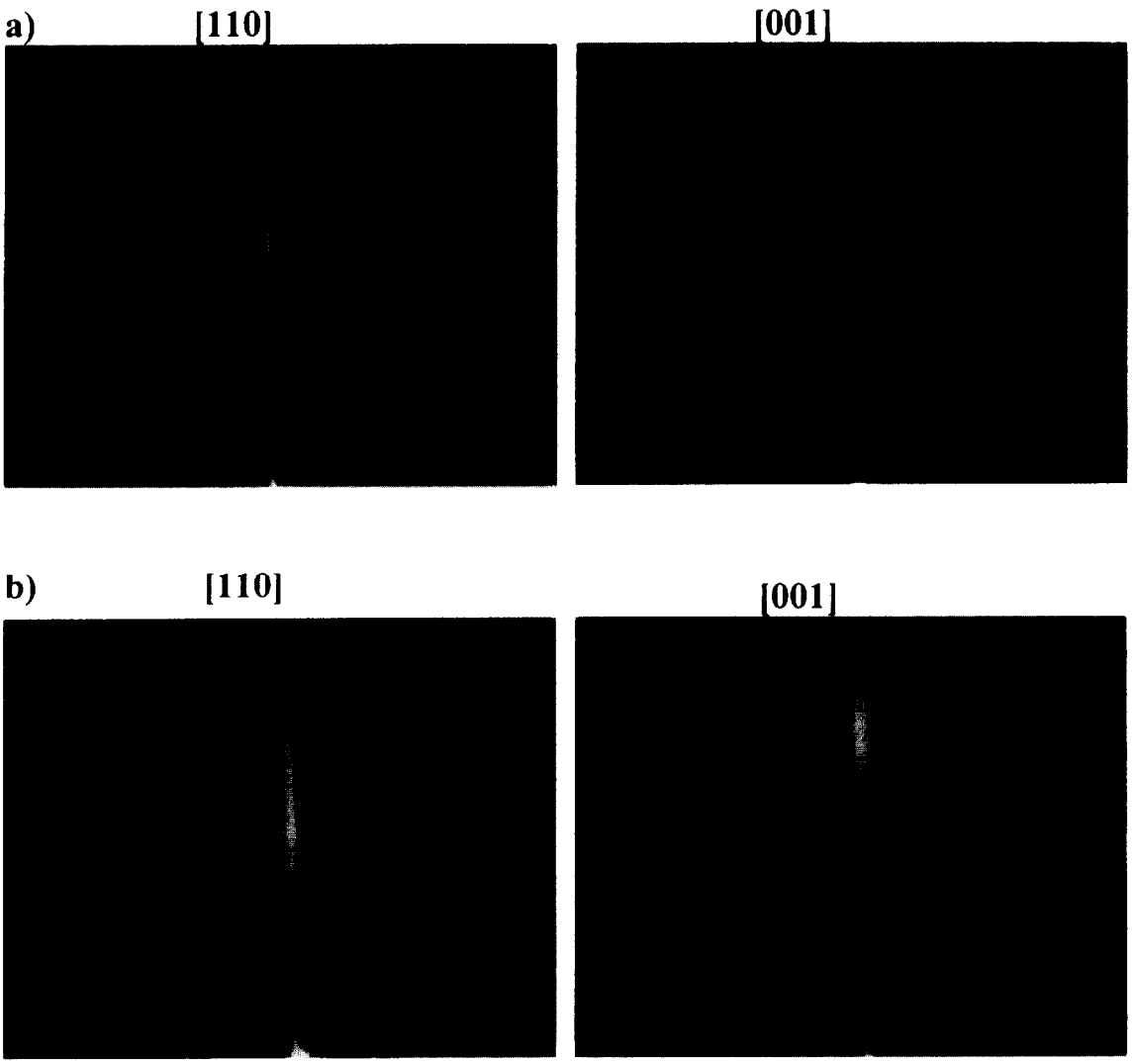

Fig. 1. RHEED patterns along [100] and [110] directions of (a) 10 nm Pt/MgO(100) and (b) 100 nm FePd/10 nm Pt/MgO(100).

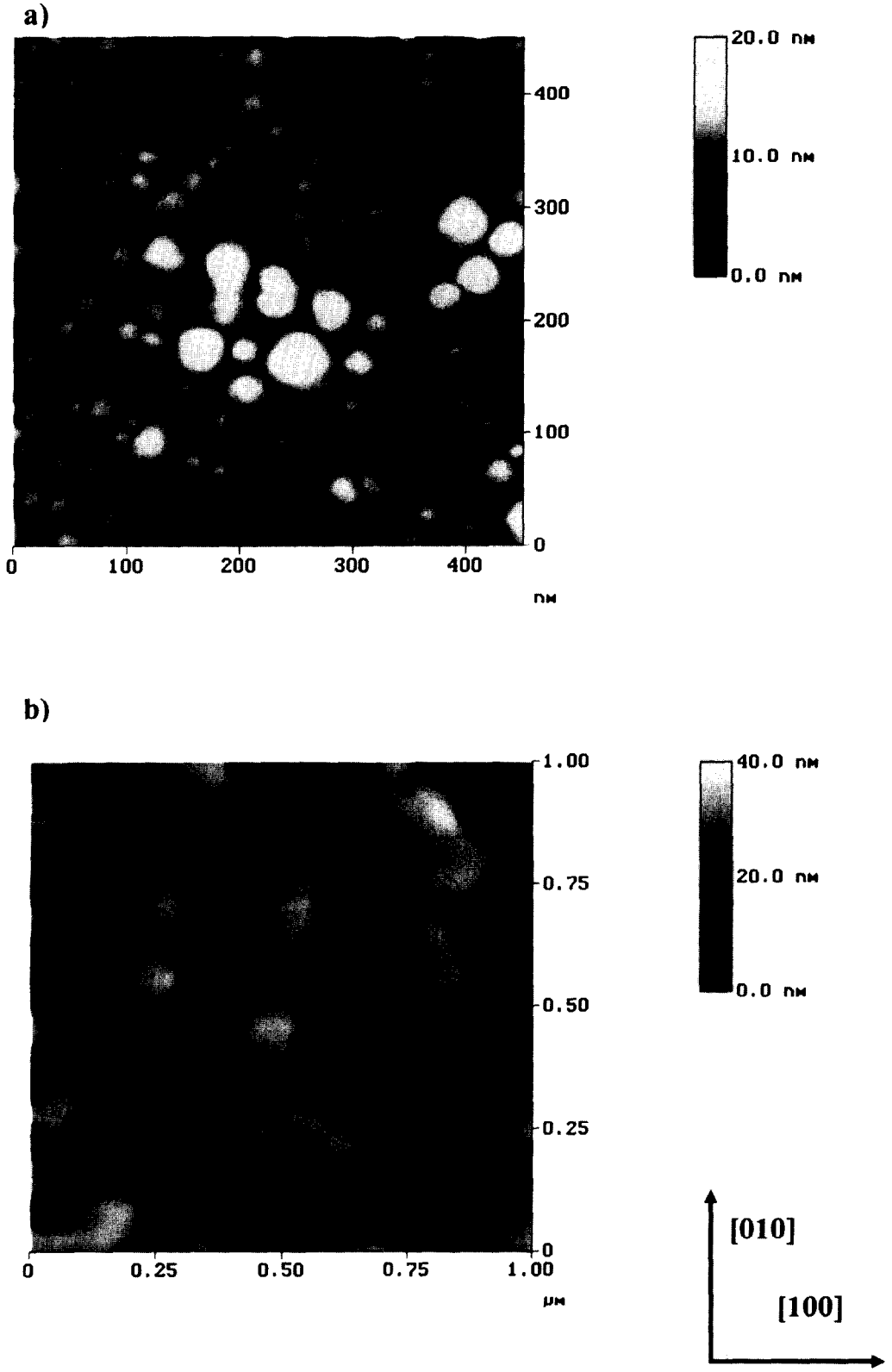

Fig. 2. Topographic AFM image (tapping mode operation) of Pt on MgO. (a) for a thin film of nominally 5 nm thickness. Notice the disparity of grain sizes. (b) For a nominal thickness of 45 nm. The resulting pattern is due to the coalescence of the individual grains shown in (a).

pendent targets both pointing to the sample. Finally, a 5 nm Pt capping layer was grown on top at room temperature to avoid oxidation of the film once exposed to air.

3. RESULTS AND DISCUSSION

Figure 1(a) shows RHEED patterns along [100] and [110] azimuths for a representative 10 nm Pt/MgO film. Sharp and intense stripes indicate the single orientation relationship adopted by the film. High quality RHEED patterns are also obtained for the FePd films grown on the Pt seeds, as shown in Fig. 1(b). It is important to note that the quality of these patterns is independent of the Pt seed thickness, therefore in all cases we have the growth of single crystalline high quality films.

Figure 2(a) shows a topography AFM image of a nominally 5 nm Pt film grown on MgO at 700°C. The Pt film is not continuous, but formed by many square shaped Pt islands aligned along the main crystallographic directions of the substrate. The sides of these islands are oriented parallel to the diagonals of the picture frame. Two kinds of Pt grains are observed: small islands of 3 nm height and 15 nm width, and bigger grains of 8 nm height and 50 nm width. The morphology of the seed layer changes when the Pt thickness is increased. In a nominally 15 nm thick Pt film, most of Pt islands are on average 190 nm wide and 27 nm high, coexisting with a few small islands. Nevertheless, we do not observe complete continuity of the film, the percentage of covered surface being about 70%. With increasing Pt thickness, a 30 nm Pt film shows even bigger islands and more coalescence. The average island side size is 290 nm and the height of these islands of the order of 38 nm. The surface of this film is very flat and about 87% of the substrate is covered by Pt. Further deposition of Pt produces almost full coverage of the MgO, with very large flat areas. Figure 2(b) shows an AFM image of a 45 nm Pt film that covers 95% of the substrate surface and is 42 nm high on average.

Figure 3 shows symmetric XRD scans for three different FePd samples all grown at 500°C onto Pt seed layers of 5, 15 and 45 nm thickness. Important changes are observed correlated with the thickness of the Pt film. In the sample grown on 5 nm Pt, both MgO(200) and FePd(002) peaks are observed, together with a broad shoulder about the region where the Pt(200) is expected. The lattice parameters of the bulk ordered FePd phase are $c = 0.3731$ nm and $a = 0.385$ nm, neither of which corresponds to the 0.3792 nm deduced from the position of the FePd peak. The presence of a FePd(001) peak is only possible if chemical order is obtained. The observation of a weak and broad (001) peak indicates a small degree of chemical order in this sample.

In the scan corresponding to the sample grown on a nominally 15 nm Pt film, two important changes are present. Now the Pt(200) is clearly observed, but giving a vertical spacing of 0.3894 nm, slightly different from the bulk value (0.392 nm). On the other hand, the FePd peaks appear shifted with respect to the peaks for the

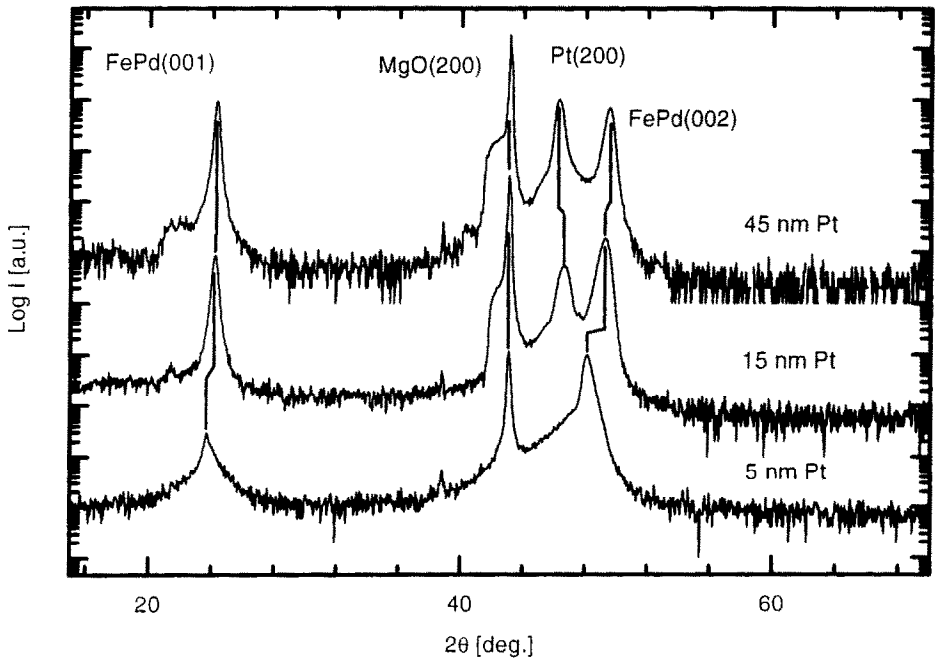

Fig. 3. Symmetric XRD scans for three FePd films grown on Pt buffer layers of different thicknesses (vertical scale is logarithmic in arbitrary units).

sample grown on 5 nm Pt, leading to a vertical spacing of 0.3686 nm, which is smaller than the bulk value. Now, the (001) peak is more intense and narrower, indicating a higher degree of ordering. Finally, the scan for the sample grown on 45 nm Pt shows the three MgO, Pt and FePd peaks again. The Pt(200) is already in its bulk position and the FePd(002) has shifted even more to the shortest out of plane lattice parameters (0.3676 nm). Again, the FePd(001) peak indicates that the FePd alloys grown is chemically ordered.

Since the FePd(001) peaks are originated only by chemically ordered areas, an estimation of the size of such ordered domains can be calculated applying the Scherrer equation [15] to the width of those peaks. We obtain in this way a domain size of 34 nm for the samples grown both on 15 and 45 nm Pt and 11.5 nm for the sample grown on 5 nm Pt.

It is a well known fact from epitaxial growth of metals or semiconductors that surface morphology controls, through the density of terraces and steps, the growth kinetics. In the case of 15 nm Pt seeds and thicker, a continuous and flat (001) oriented Pt seed layer will favor the predominance of the long range order along the [001] growth direction as determined from the width of the FePd(001) peak. In the same way, we observe that a rough and discontinuous Pt seed layer, as obtained for a 5 nm of Pt, will preclude the appearance of long range order. It has been shown [1–3] that growth on MgO(100) of single oriented Pt(100) films is only possible at high temperatures and we have demonstrated in this work that it does not necessarily produce a continuous film. This shows the problem faced when trying to grow epitaxially metals on some insulators. Here we observe that also growth on MgO(100) of FePd(001) leads to a more disordered structure. Therefore, by changing the thickness of the Pt seed layer we observe a transition in the formation of short range order in the case of discontinuous Pt films to long range order for thicker and more continuous Pt films. As a consequence, the order domain distribution will determine the magnetic anisotropy of the FePd alloy. We have observed a large perpendicular magnetic anisotropy on samples grown on thick Pt seed layers and in-plane anisotropy on alloys grown on discontinuous Pt seed layers [13].

4. CONCLUSIONS

The role of a Pt seed layer at the interface between an MgO substrate and epitaxial FePd layers has been clarified by means of a detailed AFM and XRD study: in order to obtain long range order in the FePd alloy along the growth direction and consequently a large perpendicular magnetic anisotropy, the Pt seed layer must show coalescence and offer flat and large terraces on the (001) surface to promote the appropriate growth kinetics.

Acknowledgements—This work was performed under the project "Nanoestructuras Artificiales Magnéticas: sus propiedades y aplicaciones", supported by the Spanish Commission of Science and Technology. The support under the contract program "Acciones para la incorporación de doctores y tecnólogos" (MEC) through the University Autónoma de Madrid (A.C.) and of the MUTIS Program (P.C.) is gratefully acknowledged.

REFERENCES

1. Lairson, B. M., Visokay, M. R., Sinclair, R., Hagstrom, S. and Clemens, B., *Appl. Phys. Lett.*, 1992, **61**, 1390.
2. McIntyre, P. C., Maggiore, C. J. and Nastasi, M., *Mat. Res. Soc. Symp. Proc.*, 1995, **355**, 335.
3. McIntyre, P. C., Maggiore, C. J. and Nastasi, M., *J. Appl. Phys.*, 1995, **77**, 6201.
4. Harp, G. R., Weller, D., Rabedeau, T. A., Farrow, R. F. C. and Marks, R. F., *Mat. Res. Symp. Proc.*, 1993, **313**, 493.
5. Yamamoto, K., Kasukabe, Y., Takeishi, R. and Osaka, T., *J. Vac. Sci. Technol.*, 1996, **A14**(2), 327.
6. Daalderop, G. H. O., Kelly, P. J. and Schuurmans, M. F. H., *Phys. Rev. B*, 1991, **44**, 12054.
7. Harp, G. R., Weller, D., Rabedeau, T. A., Farrow, R. F. C. and Toney, M. F., *Phys. Rev. Lett.*, 1993, **71**, 2493.
8. Lairson, B. M., Visokay, M. R., Sinclair, R. and Clemens, B., *Appl. Phys. Lett.*, 1993, **62**, 639.
9. Lairson, B. M. and Clemens, B. M., *Appl. Phys. Lett.*, 1993, **63**, 1438.
10. Mitani, S., Tahanashi, K., Fujimori, H., Osawa, A. and Nakajima, H., *J. Magn. Magn. Mat.*, 1995, **148**, 163.
11. Takanashi, K., Mitani, S., Sano, M., Fujimori, H., Nakajima, H. and Osawa, A., *Appl. Phys. Lett.*, 1995, **67**, 1016.
12. Visokay, M. R. and Sinclair, R., *Appl. Phys. Lett.*, 1995, **66**, 1692.
13. Caro, P., Cebollada, A., Ravelosona, D., Briones, F., Garcia, D., Vázquez, M. and Hernando, A., *J. Appl. Phys.* 1997, **81**, 5050.
14. Ikemiya, N., Kitamura, A. and Hara, S., *J. Cryst. Growth*, 1996, **160**, 104.
15. Warren, B. E., *X-ray diffraction*. Dover Publications, New York, 1990, p. 253.