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To cite this version:

HAL Id: jpa-00243796
https://hal.science/jpa-00243796
Submitted on 1 Jan 1974

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SOME DEVELOPMENTS IN S.E.M. INSTRUMENTATION

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Résumé. — On rappelle les travaux effectués récemment à l’Université de Sussex sur les développements instrumentaux en microscopie électronique à balayage. Les appareils mis au point comportent un détecteur universel du type « channel plate » et une platine refroidie à l’hélium liquide. On décrit une nouvelle technique qui fournit en conjonction avec la microscopie à balayage des informations cristallographiques, et on montre qu’elle présente plusieurs avantages sur les techniques antérieures. On a étudié la sensibilité de détection en microanalyse par émission X sur des couches très minces, et on a trouvé égale à environ 10^{-3} monocouche par spectrométrie non dispersive et à 16^{-1} monocouche par spectrométrie non dispersive avec un détecteur commercial.

Abstract. — Work at the University of Sussex involving instrumental developments in scanning electron microscopy is reviewed. The apparatus developed includes a versatile channel plate detector and a liquid helium cooled stage. Some results obtained using this equipment are described. A new technique for obtaining crystallographic information in conjunction with scanning microscopy is described, and shown to have several advantages over previous techniques. The detection sensitivity of X-ray microprobe analysis for very thin layers has been explored and is found to be about 10^{-3} monolayers using wave-length dispersive detection and 10^{-1} monolayers using an energy-dispersive detection system commercially available for use with a scanning microscope.

1. Introduction. — This paper reviews briefly the developments in instrumentation for scanning electron microscopy which have been made by our group at the University of Sussex over the past few years. Four main topics are discussed in the following sections.

A channel plate detector has been developed for use in a variety of detection modes, and is described in section 2. A liquid helium stage has been developed for work on condensed gases, low temperature phase transitions and magnetic materials at low temperatures. Some of these results obtained are described briefly in section 3. A new technique for obtaining crystallographic information in the S.E.M. has been discovered, which we call Electron Back-Scattering Patterns (E.B.S.P.). This technique has certain advantages over other available techniques, which are described in section 4. Finally, we have explored the use of X-ray detection techniques for the detection of submonolayer films. This technique, which turns out to have an extremely good sensitivity, and can be combined with high spatial resolution, is described in section 5.

2. A versatile channel plate detector. — A channel plate is a fibre optic electron multiplier, consisting of a large number of individual channel electron multipliers,

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REvue de Physique Appliquée. — T. 9 N° 2, Mars 1974
FIG. 1. — a) Channel plate detector in its housing, suitable for mounting on a Cambridge « Stereoscan » S.E.M. b) Exploded view of channel plate detector; A: mounting screws; B: outer lid; C: central tube; D: insulating sleeve; E: shielding plate; F: collector plates; G, J: channel plate electrodes; I, H: channel plate and housing; K, L: bottom lid with input grid; M: specimen.

depending on the electrode potentials employed, the detector can be used for secondary electrons or back scattered electrons. In the secondary electron mode, the electrodes, J, G and F (Fig. 1b) are at 200 V, 1.2 kV and 1.4 kV respectively, whereas in the backscattered mode voltages of — 1.2 kV, — 200 V and 0 V are used in order to reject the low energy secondaries. The preamplifiers are at the same voltage as plate F and an optocoupler is used to decouple the output signal, which is displayed on the S.E.M. tubes.

Examples of the performance of the channel plate in the additive and subtractive backscattered mode are shown in figure 2a-f, and the appearance of the channel plate itself is shown in g and h. These examples clearly show the transition from topographic contrast in the subtractive to « atomic number » contrast in the additive mode. Examples of the use of the subtractive secondary mode for magnetic contrast can be seen in figure 5, and examples of voltage contrast in English et al. [7], figure 11. The minimum beam currents for satisfactory operation are about 10^-12 A, limited at present by the noise in the low cost F.E.T. amplifiers used in the preamplification stage. In the secondary mode, the picture quality is similar to that using the conventional photomultiplier detector. However, in the backscattered mode the picture quality is better using the channel plate detector. We attribute this improvement to the much larger solid angle of detection (approaching 2 π steradians) of the channel plate.

3. Results obtained using a liquid helium stage. — A liquid helium stage has been built for the Cambridge Stereoscan and has been described previously [9]-[10]. It has been used to study three main areas. These are (i) condensed gas deposits, (ii) the martensitic
transformation in the Na and Li, and, in conjunction with the channel plate detector, (iii) magnetic phenomena at low temperatures. These results are described in more detail elsewhere [8] and will be published subsequently; a brief outline is given here.

3.1 STUDIES OF CONDENSED GASES. — Observations of a number of condensed gases, including Xe, O₂, CO, CO₂ and D₂, have been made, with a view to understanding the morphology of cryopumped deposits. Observations in the S.E.M. are complementary to those in the T.E.M., since the morphology can be observed when the deposit is thicker. It is clear from the present study and our previous T.E.M. observations [16]-[17], that cryopumped deposits are polycrystalline and highly defective, that lower the substrate temperature, the finer the scale of the nucleation and growth, and, almost certainly, the more porous the deposit.

Pictures of xenon have been published before [9], Fig. 4). Figure 3 shows a picture of solid D₂ condensed onto a gold substrate. The substrate consisted of a 1 000 Å film of gold (left of Fig. 3b) covering an annular support. In figure 3a the D₂ (dark) covers all but the centre of the thin Au film region, and a small square which has been previously examined at higher beam current. The low atomic number D₂ obviously has a very low secondary electron coefficient, but the high brightness of the remaining area was surprising. We suspect it is an example of the Malter effect (Seiler, 1967) in the residual gas layer; this effect is the field enhanced secondary emission from the underlying metal due to the positive charging up of the surface of a thin dielectric layer on top of the metal. This and other, charging effects were frequently observed with these insulating layers. Beam induced chemical reactions were also observed, especially in CO and CO₂. In the case of CO, CO₂ was left behind on the substrate in the bombarded area, as had been observed previously in T.E.M. studies [3].

Residual gases caused problems in these experiments even though the pressure at the specimen position was measured, using a cooled quartz oscillator, to be as low as 7 × 10⁻⁸ torr. All the effects we studied below ~ 30 K (the condensation temperature of N₂ and CO) were adversely affected. This is almost certainly due to the small escape depth of secondary electrons (10-50 Å) and the highly irregular nature of the residual gas deposit. Further examples of these problems are indicated below.

3.2 THE MARTENSITIC TRANSFORMATION IN LITHIUM AND SODIUM. — Both Li and Na transform to close-packed structures at low temperatures, and the liquid helium stage has been used to observe these transformations directly in an S.E.M. for the first time. An example of a lithium sample is shown in figure 4. On cooling below 47 K the surface markings characteristic of a martensitic transformation occurred (Fig. 4b) and the specimen could be observed to revert on heating through 95 K (Fig. 4c), new markings appearing on re-cooling (Fig. 4d). Problems with residual gas were encountered below 30 K; figure 4e shows the appearance after 15 min at 20 K. Beam-induced chemical reactions with the surface were also observed, though their nature remains uncertain.

3.3 MAGNETIC PHENOMENA AT LOW TEMPERATURES. — Magnetic materials have been examined at low temperatures, using the channel plate detector in the subtractive secondary mode. Both polycrystalline cobalt and magnetic tape samples show increased magnetic contrast with decreasing temperature down to about 100 K, due to the increased magnetic anisotropy at low temperatures. This effect is seen in figure 5a and b. However, below 30 K, we again had problems with the residual gases. We therefore did experiments in which gases were deliberately leaked into the
FIG. 3. — a) Solid D$_2$ on a gold substrate at 6.5 K. b) The same area after heating to 50 K. Both the dark area on a) — solid D$_2$ — and the bright areas on a) — condensed residual gas — have disappeared.

FIG. 4. — Martensitic transformation in Li. a) 110 K; b) 20 K; c) 160 K; d) 18 K; e) 20 K after 15 min observation.

FIG. 5. — Magnetic contrast at low temperatures in magnetic tape (stripe width ~ 100 µm). a) Room temperature; b) 73 K; c) 1 000 Å of CO condensed on the surface at < 30 K.
chamber in order to establish the thickness of the gas film which obscures the magnetic contrast. In the case shown in figure 5c, 1,000 Å of CO (or other gases) in sufficient to do this very effectively, despite the fact that the magnetic leakage field must extend away from the sample surface for about the stripe width of ~100 μm. We attribute this extreme sensitivity to the presence of large randomising electric fields near the surface of the thin, porous, irregular, residual gas deposits.

Experiments on superconductors Pb and Nb were also attempted, but without success. We feel that future experiments of this type really need U.H.V. conditions to prevent the strong residual gas effects, which have been observed, from occurring. Also for further studies of condensed gases and martensitic transformations, it would be most desirable to use additional chemical and crystallographic analysis techniques which were not available during these experiments.

4. Electron back-scattering patterns. — A new technique has been developed for obtaining crystallographic information in the S.E.M. The technique consists simply of observing the angular distribution of back-scattered electrons on a screen in the specimen chamber; the patterns so obtained contain crystallographic features which have been observed before [1] and were termed high angle Kikuchi patterns. Because the patterns arise from the back-scattered electrons we have called them electron back-scattering patterns [18].

E. B. S. P.'s have several features which are interesting in comparison with the normally used selected area channelling patterns (S. A. C. P.). Firstly, they are extremely simple to obtain; one just uses a stationary beam (spot mode) with no change of apertures or focussing. Secondly, very wide angle patterns can be obtained so that there is no possibility of mistaking the orientation. Thirdly, the size of the region from which the information comes is limited only by the cascade size in the material (0.5-1 μm diameter). Fourthly, large beam currents can be put into this spot, because the angular divergence of the incident beam is not a limiting factor. The main disadvantages at present are the need to use a tilted specimen (≥60° to the beam) and the need to know the specimen height (to about 0.5 mm) for absolute orientation determination to 1-2°.

E. B. S. P. 's have been obtained from various crystals using closed-circuit T.V. to view a simple fluorescent screen. The transmission screen, 30 mm in diameter, was fitted in the back right-hand 45° port of our Cambridge « Stereoscan », and placed about 30 mm from the specimen. An E. B. S. P. from an (001) tungsten single crystal is shown in figure 6, where the T.V. screen has been photographed with a 35 mm camera. This pattern spans an angular range of about 60° and shows the whole unit stereographic triangle. The [001] pole is clearly visible near the top centre, [011] bottom right and [111] at the extreme bottom left. Figure 6 was taken with the specimen turned to face the screen, and in this case the brightness of the pattern is rather uniform. When the specimen is not turned towards the screen the pattern shades considerably due to the approximately cosine distribution of emitted electrons. Figure 7 shows the region near the [111] pole in a pattern from the same crystal in the normal specimen orientation. The pattern is bounded on the right-hand side by the edge of the screen and shades away on the left-hand side as the shadow edge of the specimen is approached. If the detecting screen were placed in the position normally occupied by the photomultiplier detector this shading would be absent.

Patterns have also been obtained from silicon and aluminium, which demonstrates that the technique may be used for low atomic number materials. An aluminium specimen with a 5 μm sub-grain size was also used to show that sharp patterns could be obtained from individual small sub-grains, which were misoriented by about 1-2°. Using a tungsten hairpin filament in
normal « Stereoscan » working conditions one might expect to be able to put about $3 \times 10^{-7}$ A into a 1 µm diameter spot. Even with this simple fluorescent screen arrangement used for figures 6 and 7 this current is sufficient to take patterns with an exposure of only 0.25 s., and video-tape recording could easily have been used; the method is therefore very suitable for fast recording of crystallographic orientation and of dynamic observation of changes in orientation.

In order to measure specimen orientations absolutely to about 1-2° accuracy, it is necessary to know the specimen height and the specimen-screen distance to within about 0.5 mm. The determination of orientation is most easily accomplished by measuring the coordinates of a prominent zone axis on the screen and the angle at which a plane (which goes through this zone axis) crosses one of the coordinate axes. For example, in figure 7, the $x$, $y$ position of the $[111]$ pole and the angle, $\phi$, of the almost horizontal $(121)$ plane to the $y$ axis would be suitable input data. A programme has been written for a PDP-8E computer to convert this data into the specimen orientation. Alternative versions could be constructed which used, for example, the coordinates of 3 poles to fix the specimen orientation. Such a measurement could, in principle, obviate the need to know the specimen height, as it could be determined self-consistently by the data points. Care has to be taken, however, as the positions of the poles often have to be known with great precision for such an approach to work.

Experiments are underway to observe the patterns directly using a T. V. camera which is fibre-optically coupled to a transmission screen in the specimen chamber. When the S. E. M. itself is switched to the spot mode, to obtain the E. B. S. P., the S. E. M. scan generator is used to drive the T. V. camera the second video tube and the record tube, leaving the S. E. M. beam and the first video tube in the spot mode. Thus the signal from the T. V. can be displayed and recorded using the normal S. E. M. facilities. The fibre optic coupling is much more efficient than the system used to record figures 6 and 7. A reduction of a factor of 100 in the beam current required should be obtainable.

This type of recording system, though not yet proven, should be extremely useful not only for E. B. S. P.'s but for transmission diffraction and reflection diffraction patterns as well. In contrast to the scanning method of obtaining a diffraction pattern, where either the incident or the transmitted beam is rocked and the signal is collected behind a small aperture [19], the T. V. camera method should be extremely efficient. This is because the whole of the pattern is collected all the time by the face plate of the T. V. tube, rather than being thrown away for almost all the time in the scanning method. It should certainly be possible to obtain all three types of diffraction pattern at current levels approaching that used for obtaining high resolution scanning micrographs, with very short exposure times.

5. Submonolayer detection by X-ray analysis. — In this last section we describe a technique developed by one of us [5] to detect very small quantities of material on thin film substrates by X-ray microprobe analysis. In conventional X-ray microprobe analysis of bulk samples, the minimum amount of material detectable corresponds to about one monolayer, i.e. $\sim 10^{15}$ at/cm$^2$. However if a thin film, particularly of a material with a low atomic number, is used as the substrate, the white X-ray background from the substrate is greatly reduced. In the work described here, detection sensitivities of $10^{-3}$ monolayers ($\sim 10^{12}$ at/cm$^2$) have been achieved for a range of metals Al, Ti, Ag and Au on 200 Å thick carbon substrates, by using wavelength dispersive analysis (WDX) in a combined electron microscope-microprobe analyser (EMMA). Energy dispersive analysis (EDX) in the Stereoscan S. E. M. yielded a limit of about $10^{-1}$ monolayers.

Figure 8 shows the relation obtained for the metals examined in EMMA at 60 kV, between the X-ray intensity and the deposit thickness of evaporated films. The linear relation extends from $10^{-3}$ monolayers ($\sim 3 \times 10^{-3}$ Å) and $10^{-2}$ monolayers ($\sim 3 \times 10^{2}$ Å). It is clear that neither the intensity of the X-rays nor the detection limit of the method, is a strong function of the atomic number, $Z$, of the material analysed. This is because at high $Z$ one uses the $M_{z}$ line and at low $Z$ the $K_{x}$ line, and this tends to cancel any intrinsic $Z$-dependence of the X-ray emission.

This application of X-ray analysis is an example where the superior signal/background (S/B) ratio of the WDX technique wins over the superior count rate of the EDX technique. The S/B ratio of the WDX is about a factor of twenty better at the same energy,
due to the fact that the energy resolution of EDX is \( \sim 150 \) eV whereas WDX can approach 5 eV. In addition, overlapping peaks due to contaminants in the system can cause problems for EDX but not for WDX. The WDX signal is however very small. At the lowest levels shown on figure 8, a 2 \( \mu \)A beam current was used in a 2 \( \mu \)m diameter spot for 400 s counting time. Thus the signal intensity (P-B) corresponds to a total count only 55 X-ray quanta for the lowest Ti point shown, whereas the background intensity was 80 counts; this accounts for the large counting error near the lower level of detection.

In EMMA more than 50 \% of the background signal does not come from the substrate, but from X-rays produced by scattered electrons. In a typical scanning microscope, where the facilities for trapping scattered electrons are much less sophisticated than in EMMA or non-existent, this fraction must be much higher, although the energy range available in S.E.M.'s (10-30kV) gives the highest S/B ratios. Thus we expect the use of WDX in conjunction with commercial S.E.M. will result in a lower limit of detection which is intermediate between that obtained for the WDX and EDX techniques in the present work.

In any case, this work has demonstrated that X-ray microprobe analysis of material on thin film substrates has a comparable sensitivity to other analysis techniques such as Auger electron spectroscopy (A.E.S.) [4], low energy ion scattering [2] or autoradiography [14]. In addition the method can be used with 2 \( \mu \)m spatial resolution so that a minimum of order 10\(^5\) atoms can be detected. Unlike the first two techniques mentioned, the method is not sensitive to whether the atoms are at the surface of the substrate, and this means that the vacuum conditions are less critical. When used in conjunction with the line scan mode of the S.E.M. to display spatial variations of concentrations at the monolayer level, the method would be easier to use than A.E.S. as it has been reported that computer control is required [12]-[13] even just to detect the presence or absence of a given element in the Auger signal. We can, however, expect further development of analogue A.E.S. line scan and area mapping techniques which will make the X-ray and A.E.S. detection of monolayers of comparable sensitivity. Any such techniques would, however, be very time consuming, and area mapping would need to use a very coarse raster to display pictures of concentration variations at the monolayer level.

Acknowledgments. — One of us (J. A. V.) was very grateful to Dr. Fontaine for the invitation to visit Lyon and to the Marseille group for their hospitality during the summer term of 1973. We are most grateful to Tube Investments Research Laboratories for the use of EMMA, and in particular to Dr. C. J. Cooke and M. P. D. Hunneyball for their advice and help. We are also grateful to the support from AERE, Harwell (K. H. E.) and SRC (B. W. G. and J. A. V.) which enabled this work to be done.

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