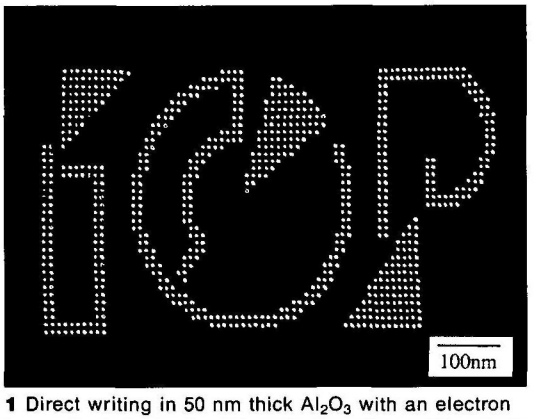
Advanced materials

Wavelength imposes a fundamental limit on photon-based lithographic techniques whereas electron beams now offer resolutions hundreds of times better

Ultimate limits of lithography

CHRIS MORGAN, GIIN SHAN CHEN, CHRIS BOOTHROYD, SIMON BAILEY AND COLIN HUMPHREYS

IN 1959 the famous bongo player and Nobel laureate Richard Feynman first posed the question: "Why can't we write the *Encyclopedia Britannica* on a pinhead?". If an electron beam could be focused to a spot only one atom in diameter, he reasoned, then it might interact with individual atoms on a surface and writing on an atomic scale would be possible. Aberrations in electron lenses prevented electron beams being focused to spots with diameters less than ~2 nm in 1960. However, recent advances in electron optics have made it possible to focus electron beams to less than 0.5 nm in diameter, and at Cambridge we have used a focused electron beam to write a portion of the *Encyclopedia Britannica* sufficiently small to demonstrate that the entire encyclopedia could indeed be put on a pinhead. We have used the same technique, known as SCRIBE, to write the Institute of Physics logo by cutting dots through a piece of amorphous aluminium oxide (Al_2O_3) 50 nm thick (figure 1). Each dot has a diameter of 5 nm (50 times smaller than currently possible with conventional optical lithography) and took 0.01 s to produce.



1 Direct writing in 50 nm thick Al_2O_3 with an electron beam. Each dot is about 5 nm in diameter and the whole image took about 10 s to write using a scanning transmission electron microscope

The main forces driving lithography to smaller dimensions are the science push of novel physics and the market pull of small electronic devices. Advances in lithography have enabled dramatic progress to be made in microelectronics. In 1960, for example, there was one device on a silicon chip – now there can be over 50 million. No single factor has had such a profound effect on modern society as the continuing development of microtechnology. The processing power made available by the progressive miniaturisation of electronic devices has led us into the current age of information technology.

Lithographic options

Optical The production of miniature devices has long been the preserve of optical (i.e. visible or ultraviolet light) lithography (see box 1). A photosensitive resist is exposed through a mask and then developed, leaving certain regions of the substrate exposed for whatever form of etching, doping or deposition is required. This technique has been (and will continue to be) preferred for the fabrication of devices with features of the order of 0.25 μm or greater since the short exposure time and large area which can be exposed in one step combine to give a very high throughput.

X-ray The best and cheapest way to routinely obtain 0.2 μm lithography is a key question facing the semiconductor industry as 0.2 μm lithography is required for the next generation of 256 Mbit computer memory chips. The minimum feature size cannot be much less than the wavelength of the light used to expose the resist, which for UV light is about 0.2 μm. Excimer lasers have to be used for 0.25 μm optical lithography but breaking the 0.2 μm barrier optically will need innovative resists, masks and optics. X-rays have a much shorter wavelength (down to 0.1 nm) and potentially much better resolution but, as yet, there is no lens that can adequately focus X-rays. Some scientists expected X-ray lithography to displace optical lithography below 0.25 μm but optical lithography keeps advancing! The ultimate resolution achievable with X-ray lithography is limited to about 0.1 μm by Fresnel diffraction effects from the mask (since X-rays cannot be focused) and by photoelectron track lengths in the resist. The ultimate resolution achievable using optical lithography is also probably about 0.1 μm. Whether X-ray lithography or optical lithography will be the best and cheapest technique in the 0.1–0.2 μm range is still an open question of considerable financial importance to the microelectronics industry.

Enter electrons

Electrons accelerated through 100 kV have de Broglie wavelengths of about 0.004 nm and can be focused with magnetic lenses; however, the main problem with electron beam lithography (EBL) is throughput. Whereas optical and X-ray lithography methods expose large areas of semiconductor wafers simultaneously, most EBL methods

irradiate the wafer sequentially, with the beam scanning across the wafer in the desired pattern. Thus optical lithography is the best technique for the mass production of devices down to 0.25 μm resolution. Electron beam lithography comes into its own for low-volume ASICs (application specific integrated circuits) and for all lithography requiring a resolution of better than 0.1 μm.

Conventional electron beam lithography uses resists such as polymethylmethacrylate (PMMA). The incident electron beam damages the resist (the primary damage process being ionisation which then leads to bond breaking) and the damaged region is then removed by a solvent. To achieve a reasonable speed of writing it is obviously desirable to have as high a beam current density as possible. Many EBL systems use lanthanum hexaboride (LaB_6) electron guns from which focused probes 5 nm in diameter can be produced. Although field-emission sources can produce smaller probes with higher beam current densities they have not been widely used for lithography due to stability and noise problems.

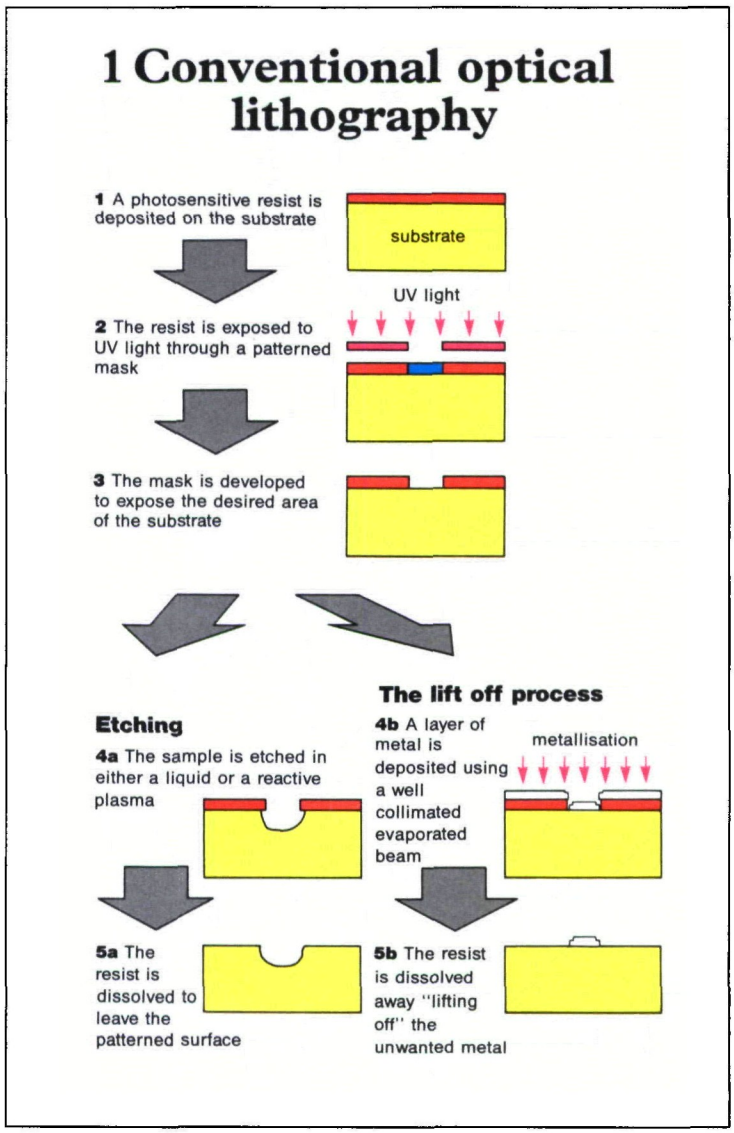
The beam can have either a finely focused Gaussian profile, which gives the best resolution but is rather slow, or a square profile (produced by passing the beam through a pair of overlapping square apertures) which allows far faster pattern transfer but is less flexible for producing complex patterns. There are two ways of writing a pattern with electron beams: in the scan method the beam is rastered over the substrate and blanked or unblanked to form an image made up from pixels; in the vector method the beam is directed at only those portions of the chip which require exposure. The vector method allows much greater data compression and speed.

Electron beam lithography employing PMMA has been successfully used to create structures with line widths down to 10 nm; these include ultra-small transistors, low-dimensional structures to investigate quantum electron effects, and X-ray zone plates. The minimum spacing of features obtainable using conventional EBL is normally limited by "primary" electrons which, having passed through the resist and written the desired pattern, are back-scattered in the semiconductor substrate by multiple phonon scattering. These electrons pass back through the resist and expose areas they were not intended to. If the substrate is thick, these back-scattered electrons can limit the resolution to ~1 μm. However, if thin substrates and higher energy electron beams are used most of the electrons will pass through the substrate and will not be back-scattered. Various factors may then limit the ultimate resolution in conventional EBL: "secondary" electrons generated in the resist by the primary electrons and travelling perpendicular to the primary beam direction have a range of ~5 nm. Ionisation of the resist "at a distance" by Coulomb interaction with the primary electron beam is also a problem. Classical impact parameter considerations (or the uncertainty principle) show that an incident electron beam can transfer sufficient energy to an atom to excite an atomic electron and hence "expose" the resist at a distance from the primary electron of up to ~5 nm. A third factor is the size of the PMMA molecules (~2 nm) which imposes a limit ~4 nm. The overall result of these effects is to limit the ultimate resolution of conventional electron beam lithography to about 10 nm.

Lithography without resists

In 1978 Alex Broers and co-workers at IBM found that they could form holes only 5 nm in diameter directly in NaCl using a focused electron beam from a LaB_6 source without a resist. The writing in NaCl was not permanent and the holes closed up due to attack by atmospheric water vapour. The first permanent writing on a nanometre scale was performed by Margaret Mochel and colleagues at the University of Illinois,US, in the ceramic material sodium β-alumina in 1982. They used 100 keV electrons from a field-emission gun to cut 2 nm holes through 100 nm thick materials and demonstrated that the mechanisms generating the holes were more complex than simple vaporisation by the electrons.

Subsequent research has shown that a focused intense electron beam of energy typically 100 keV can cut through a wide variety of materials, for example Al_2O_3, MgO, Si, Al, AlF_3, etc. The minimum hole diameter or line width produced so far is about 0.5 nm, but in principle it would seem to be possible to remove a single column of atoms, given a sufficiently finely focused electron beam. This technique has been given the acronym SCRIBE (sub-nanometre cutting and ruling by an intense beam of electrons) and does not involve any resists or chemical development. The electron beam can cut through 200 nm or more of material and lines can be produced by moving the



2 Resist exposure mechanisms

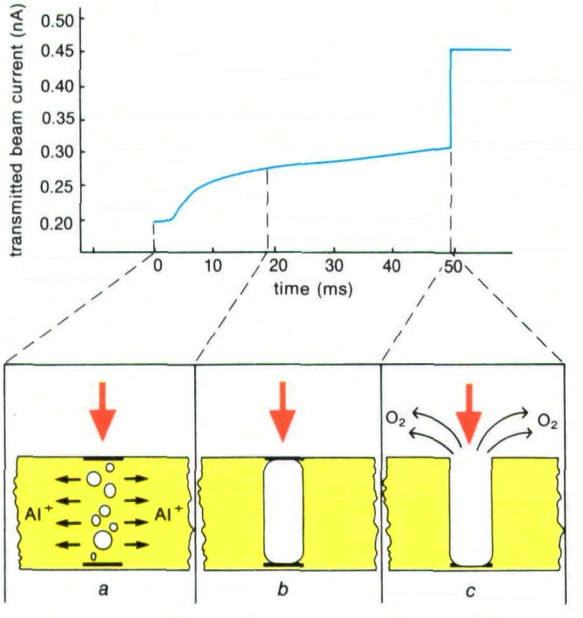
The mechanisms by which material is lost from inorganic resists on exposure to an intense electron beam are varied and intriguing. We consider aluminium oxide as an example. It is possible to drill holes 2 nm in diameter in amorphous alumina films in less than 20 ms using an intense electron beam. Evidence for the hole-formation mechanism comes from measuring the electron beam current transmitted through the specimen as a function of time (left) and from observing the film during irradiation.

The amorphous aluminium oxide structure is probably disrupted by a multiple ionisation process first suggested by Knotek and Feibelman (*Surf. Sci.* 40 78 (1979)). In this an incident high-energy electron ionises an Al^{3+} ion by removing an electron from its highest occupied level (2p). This hole is then filled by an electron from the (higher) 2p level of an adjacent O^{2-} ion in an interatomic Auger process.

The excess energy of the system (the energy difference between the 2p levels of O^{2-} and Al^{3+}) can be released by the emission of one or two Auger electrons from the oxygen ion. The original O^{2-} ion therefore becomes either O or O^{+} and is thus repelled from the aluminium oxide – either to the surface, from which it is desorbed, or to collect in bubbles in the interior (top right).

It has been suggested that a high radial electric field is induced in the specimen (as a result of ionisation by the incident beam) and that this field separates charged ions in the disrupted structure. Electrostatic calculations predict that if the sample is exposed to a "top-hat" beam, the greatest electric field exists around the edge of the beam – this is the region which damages first (bottom right). The top-hat intensity profile is produced by passing the electron beam through a circular aperture and the damage pattern even reproduces the non-uniform edge of the aperture.

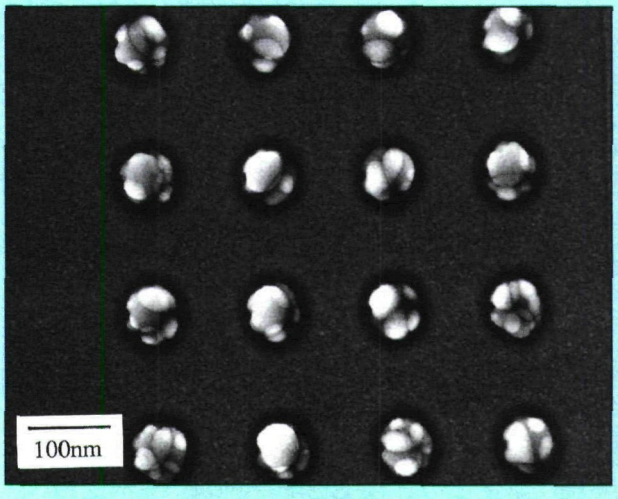
This is just one example of the sort of complex mechanism which can occur when inorganic resists are exposed to intense electron beams. In contrast, mass loss in magnesium oxide is thought to be due to sputtering of MgO "molecules" from the electron "exit surface" of the material. In zinc oxide the oxygen is preferentially desorbed to leave a plug of metal which can then be removed by further exposure if required. Aluminium is proposed to damage by the migration of voids from the electron exit surface to the electron entrance surface by a sputtering process. The hole-formation mechanism for silicon is still not clear.

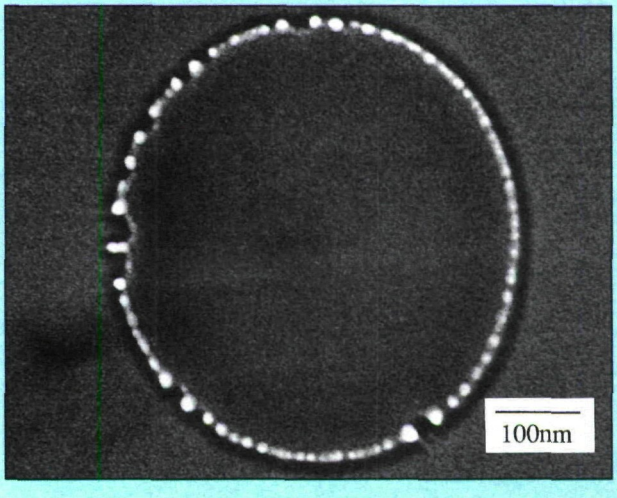


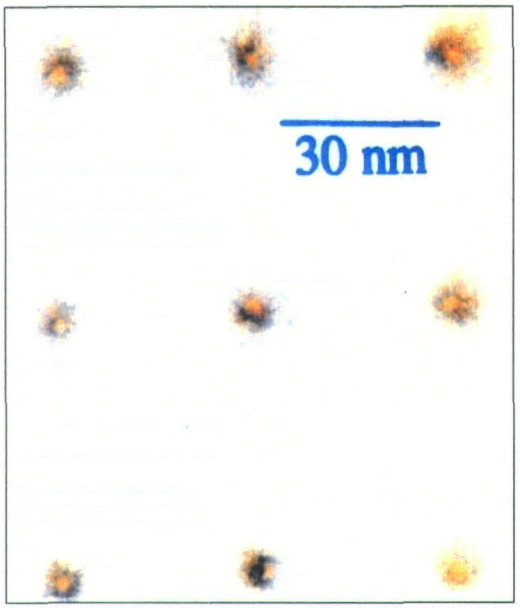
(*a*) Immediately upon irradiation oxygen is desorbed from the surface to leave a "capping layer" of aluminium. Within the bulk, oxygen ions are displaced to form bubbles. Positive aluminium ions are swept radially away from the beam by the electric field caused by charging under the beam.

(*b*) Further irradiation causes the bubbles of oxygen to grow, and then to merge into one large bubble.

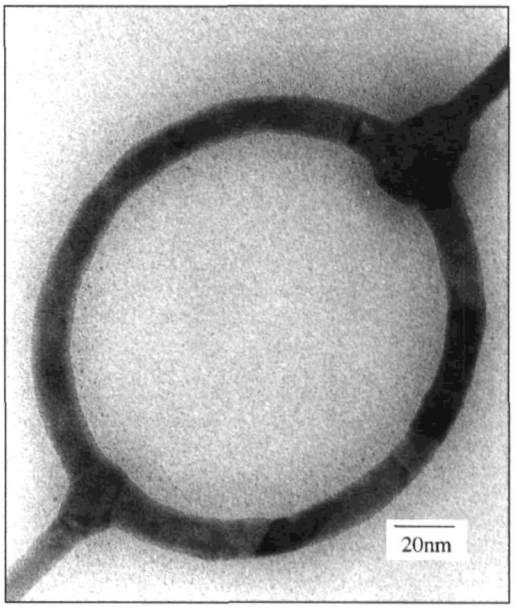
(*c*) Finally the pressure of oxygen gas within the bubble causes the bubble to burst through either one or both of the capping layers







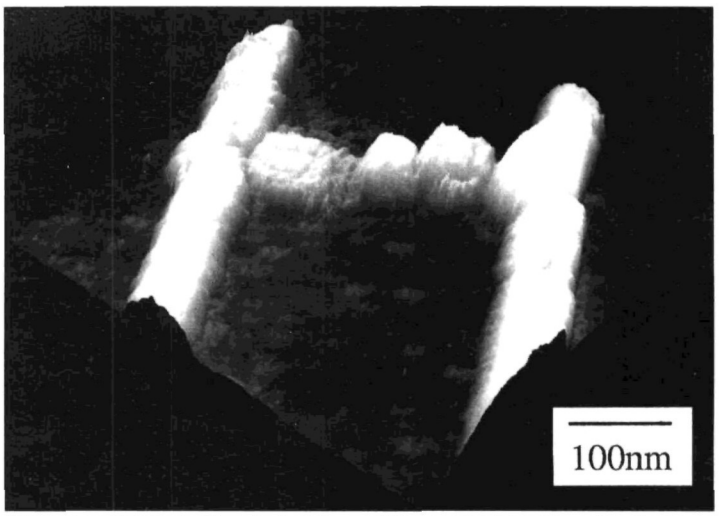
◀ **2** Quantum dots of silicon formed directly in 15 nm thick SiO_2 by an electron beam. This figure is a combination of a Si (yellow) and a SiO_2 (blue) plasmon loss image. Plasmon loss images are made using electrons which have lost energy by creating a plasmon in the sample. The Si dots appear yellow and the unexposed SiO_2 film appears white because the tails of the broad SiO_2 plasmon loss peak overlap the narrow Si plasmon peak



▶ **3** Gold–palladium ring with a wire width of 12.5 nm fabricated on silicon nitride in a 400 kV transmission electron microscope using contamination resists followed by ion-milling (Courtesy A N Broers, Cambridge)

beam. Some of the different mechanisms at work in the cutting process are explained in box 2.

One application of the SCRIBE technique is the fabrication of silicon quantum dots and wires direct from SiO_2, which has been demonstrated at Cambridge University. If SiO_2 is irradiated by an intense focused electron beam, both oxygen and silicon are removed, but oxygen is removed faster. A cylinder of silicon, typically 2 nm in diameter, is formed in the centre of the irradiated area surrounded by an annular region of SiO_{2-x} (figure 2 shows an array of such quantum cylinders). If the beam is moved a quantum wire is formed. These structures are significantly smaller than those produced using conventional EBL and quantum confinement effects may result in these structures being light emitting. If light emission can be achieved in a controlled manner from silicon this may have important applications (see "Silicon optoelectronics at the end of the rainbow" by Leigh Canham *Physics World* March 1992 p41).



4 Two lines fabricated by electron beam lithography joined together by a row of dots formed in a scanning tunnelling microscope (Courtesy M Welland, Cambridge)

Contamination resists

Hydrocarbon contamination in electron-optical instruments is frequently a nuisance. An intense electron beam can cause the "cracking" of hydrocarbons near, or on, the sample surface and leave a carbonaceous layer on the sample. This is a problem for analytical microscopists but has been successfully exploited for lithography. Lines of contamination may be deliberately deposited on the substrate by a focused electron beam and used as a negative resist. Although the exposure dose required is too great to allow a practical speed of writing, it is a useful and flexible technique for creating research structures with line widths of around 10 nm and has been successfully used to fabricate Fresnel zone plates for use as X-ray lenses. A combination of contamination resists and ion-milling were used to produce the gold–palladium ring shown in figure 3 – the wire width is 12.5 nm.

STM

Scanning tunnelling microscopy (STM) may be the ultimate tool for nanofabrication with atomic resolution. In imaging mode a very sharp tip is scanned across the sample surface in a raster pattern with the tip about 1 nm above the specimen surface. The tunnelling current between the tip and the (conducting) substrate is monitored and the vertical position of the tip is altered to maintain this constant. The resultant map of tip displacements reveals the surface topography (although care must be taken that it does not in fact reveal the tip topography!).

There are several methods of employing the STM for nanofabrication. The simplest is direct tip-to-surface contact, either punching a hole in the substrate or leaving some of the tip behind. The STM can also be used to deposit matter reactively from the surrounding atmosphere by applying an appropriately high bias pulse between the tip and the surface. The ultimate resolution achieved in the STM was demonstrated by Don Eigler and Scott Schweitzer who, using variable tip biases, successfully manipulated individual adsorbed xenon atoms to form the letters IBM on a substrate. Figure 4 shows two lines 280 nm apart, produced by electron beam lithography and joined by a row of dots, formed using the STM.

Various groups worldwide are working to combine the best features of STM and electron beam lithography. A novel example is the work of Philip Chang and colleagues at IBM who have built a miniature EBL system using an STM tip as the field-emission source and microlenses to form a miniature electron "optical" column. The microlenses have a bore of just 1 μm and are themselves made

using electron beam lithography. The microcolumn includes the STM source, microlenses and a scanning system, and is only 2.5 mm high. It is proposed to construct an array of such microcolumns to achieve high-throughput sub-100 nm electron beam lithography in a routine manner to fabricate future microelectronic circuits.

The future

It is clear that improvements in lithography hold the key to many exciting future developments in physics, electronics and beyond. The potential to routinely produce structures ten times smaller than currently available will greatly increase the power of semiconductor technology. If structures a hundred or a thousand times smaller can be produced a new breed of quantum device may be possible, together with novel structured catalytic surfaces for chemistry, and new X-ray and electron optical structures. Nanometre-scale lithography will also be increasingly used to develop nanostructures for biological and medical applications.

Further reading

A N Broers 1988 Limits of thin film microfabrication *Proc. Roy. Soc.* **A416** 1

R P Feynman 1960 There's plenty of room at the bottom in *Miniaturisation* ed H D Gilbert (Chapman and Hall, New York and London) p282

C J Humphreys 1989 Materials science and engineering in Britain *Angewandte Chemie Advanced Materials* **101** 1103

C J Humphreys *et al.* 1985 Nanometre-scale electron beam lithography in *Electron Microscopy and Analysis 1985* ed G J Tatlock (IOP Publishing, Bristol) p1

C J Humphreys *et al.* 1991 100 keV electron beam damage of metals, ceramics and semiconductors – implications for micro-analysis and nanolithography in *Electron Microscopy and Analysis 1991* ed F J Humphreys (IOP Publishing, Bristol) p319

S D Berger *et al.* 1987 Electron energy-loss spectroscopy studies in alumina produced by intense electron-beam irradiation *Phil. Mag.* **B55** 341

Chris Morgan, Giin Shan Chen, Chris Boothroyd, Simon Bailey and **Colin Humphreys** are in the Department of Materials Science and Metallurgy, University of Cambridge, Cambridge CB2 3QZ, UK

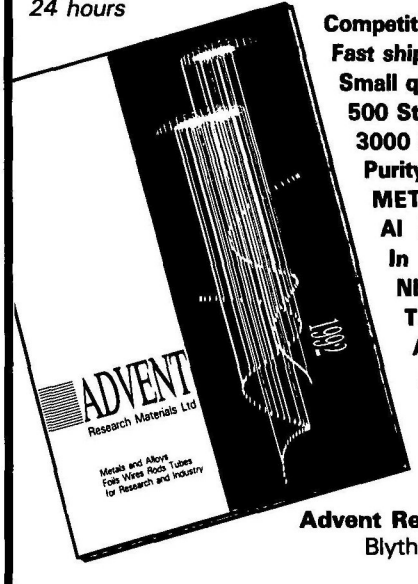
Correction

Flat panel displays or bust?
by Kristina Johnson
(Physics World, September 1992 pp37–42)

The author has requested that the following corrections be made to the published text:

- p37, third paragraph. The fourth and fifth sentences should be reversed, i.e., the substrates are rotated with respect to one another before the liquid crystal is flowed into the cell cavity at elevated temperatures under vacuum such that the alignment layers are rubbed in the perpendicular direction.
- p40, third paragraph, last sentence. The contrast ratio should be 12:1 and the turn-on times should be 120 μs.

GOLD AND PLATINUM
PRODUCTS FOR RESEARCH AND TECHNOLOGY
As specialists in gold and platinum products whatever your needs – however urgent – call 021-766 6022. We will be pleased to discuss your requirements.
BIRMINGHAM METAL COMPANY LIMITED
SPECIAL PRODUCTS DIVISION
Garrison Street, Bordesley, Birmingham B9 4BN.
Telephone: 021-766 6022.
Fax: 021-766 7485.
INSERT 14 ON REPLY CARD

Metals & Alloys
Foils Wires Rods Tubes
The competitive source for all your requirements
Shipments within 24 hours
Prompt response to enquiries


Competitive prices
Fast shipment
Small quantities
500 Stock lines
3000 Standard packs
Purity 99.9% - 99.999%
METALS
Al Be Cd Co Cu Au Hf In Ir Fe Pb Mg Mo Ni Nb Pd Pt Re Rh Si Ag Ta Sn Ti W V Zn Zr
ALLOYS
Al Bi Cu Fe Mg Ni Nb Pt W
Free 80 page Catalogue
Advent Research Materials Ltd
Blyth Road Industrial Estate
Halesworth · Suffolk
England IP19 8DD
ADVENT
Telephone 0986 874555
International +44 986 874555
Fax 0986 874557
International +44 986 874557
INSERT 15 ON REPLY CARD