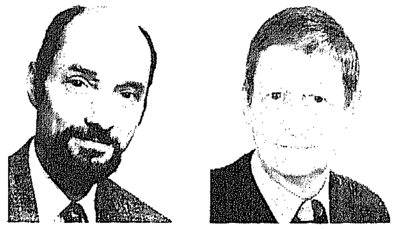
A 1000 kV TEM Running Over 25 Years



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A JEOL 1000 kV High-Voltage Electron Microscope has been running since 1971 when it was installed in Halle (Saale),Germany, with 280 thousand micrographs being exposed up to now .

1. Background

Halle (Saale) is an old university town located in the former East Germany (German Democratic Republic, GDR). It was in the late 60s when economy was rapidly growing even in a socialist country like the GDR. Governmental research funds were provided to expand the facilities and staff of the Academy of Sciences of the GDR. An institute belonging to this academy, the Institute of Solid State Physics and Electron Microscopy, had been established in Halle (Saale) in 1959 by Heinz Bethge. Owing to his activity and the scientific reputation of the institute, it was possible to raise the necessary amount of money for buying a JEOL high-voltage transmission electron microscope (HVEM), serial # 6, operated at 1000 kV. Worldwide, there were about 30 HVEMs running in the 70s — mainly in Japan, the UK, and the U.S.A., among them two in Stuttgart/Germany and one each in Stockholm/Sweden, Moscow/USSR and Halle (Saale) /GDR.

The HVEM in Halle (Saale) — shown in **Fig. 1** — was installed in a new dedicated building between 1970 and 1971. From the very beginning of constructional work, a staff of a leading scientist (one of the authors, G. K.), two technicians, and an increasing number of scientific co-workers were involved in all details. It was very stimulating to assist the Japanese team of up to five engineers in the assembly and during the test period of all parts of the big machine. Various provisional installations had to be made, including a wooden 1 m² basement for a heavy (1000 kg) motor-generator supplying the 3 kHz input of the high-voltage system. When connecting this provisional installation for the first time, all persons kept at a safe distance of several meters. Right from the beginning, the staff was involved in all details of the later operation, including the scheduled annual overhaul, according to a five-year contract with JEOL. Subsequently, the staff was able to overhaul the whole system — including the complete disassembly and cleaning of the high-voltage system and the accelerator tube — with only occasional help by JEOL. This was essential as it saved much money in convertible currency, which was very rare in the GDR. Later on, it turned out that the accelerator was working well for several years without cleaning, and the annual downtime for repair and cleaning other parts was reduced to about 3 weeks. Up to now, more than 280,000 micrographs have been exposed.

In general, it was advantageous to have an experienced team of permanent employees. Thus, a lot of difficulties could be solved which arose from the chronic shortage of materials and convertible currency typical of all socialist economies. Two examples may be mentioned: After running the microscope for about 6 years, a set of 20 spare insulators for the 1 MV accelerating vacuum tube was managed to be produced in a GDR factory according to drawings kindly provided by JEOL. It was a similar but more tedious in-house work to design and build a new 1 MV resistor column — located within the high-voltage gas-pressurized tank — as the measuring resistor to provide the feedback voltage for the 1 MV voltage control and stabilization circuit. After several trials, the 14 stages of this column were built from 4480 thin-film resistors individually pre-tested. This resistor column has been working well for 4 years.

The background changed drastically after the reunification of Germany in 1990. Since 1992, the HVEM facility has been part of the new Max Planck Institute of Microstructure Physics established in Halle with the directors of the experimental departments Ulrich Gösele and Jürgen Kirschner. The instrument has now mainly been used for *in situ* investigations. Accordingly, the responsibility has been passed to the other author (U. M.). Owing to the skill of Christian Dietzsch (physicist) and Wolfgang Greie (technician), both from the most experienced former staff, all technical problems and breakdowns have been solved without help from outside so that the microscope is still working stably most of the time. Because of the still high demand for conventional applications and, particularly, because of the unique possibilities of *in situ* experiments, it is planned to maintain the instrument also over the next few years.

2. Technical upgrade and *in situ* facilities

Because of the low budget of convertible currency available during GDR times, the HVEM was bought at a very low price level so that its technical facilities had to be upgraded and expanded in various ways. Nearly all improvements were laboratory-made in order to achieve unique facilities.

2.1. Upgrade

At the very beginning, two upgrades badly needed were done: a film numbering device (where an electromechanical display is optically exposed) and an automatic starting unit, switching on the pumps, etc. after sudden power cuts (which were quite often during construction work within the campus). This starting unit was built by using electromechanically driven switches since no IC's were available. In addition, the noisy 3 kHz transformers of the high-voltage supply were put in insulating boxes (and therefore water-cooled) to ensure sound protection.

Later on a TV system was installed, consisting of a transmission fluorescent screen, a 45° mirror, a lens-coupled TV camera, and an extension of the X-ray lead shielding.

A quite useful feature of the HVEM is the large diameter (about 300 mm) of its specimen chamber as well as the diameter of 36 mm of the upper bore of the top-entry objective pole piece. This provides enough room for attaching special cartridges and devices.

A precise universal goniometer was built in 1977 by one of the authors (G. K.) in order to improve and expand the routine facilities for using electron diffraction and diffraction contrast. First, the specimen airlock and top-entry transfer were completely redesigned to better manipulate à new standard specimen cartridge for larger samples. It may clamp specimens of up to 8 mm in diameter and 1 mm in height. This cartridge also enables double-tilting of up to 45° at all azimuth angles by means of the top-entry goniometer mechanism shown in **Fig. 2**. The cone-shaped specimen cartridge C (shown separately on the right) is kept by a cartridge holder H, which is supported by a universal joint precisely located within the specimen plane of the objective lens. At about 13 mm above this plane, a small brass ball is fixed to the cartridge holder (below the letter H). This ball is gripped between actuating tongs T, each arm coupled to a horizontal gear drive G. On moving these drives into opposite directions, the opening angle of the tongs is varied, thus tilting the cartridge in y direction. If the drives are moved synchroneously into the same direction, the tongs translates without altering the angle, thus tilting the cartridge in x direction. Tilt angle and azimuth are displayed in two dimensions on a screen in front of the operator. This screen is shown as a bright area above the viewing chamber in Fig. 1. It is connected to the vertical tilting drive rods with their handle knobs conveniently located on the left and right sides of the viewing window.

2.2. *In-situ* facilities

Together with the microscope, various top-entry attachments for *in situ* investigations were bought:

Specimen heating stage:
An airlock-exchangeable specimen cartridge can be resistance-heated up to 1070 K and tilted up to 30° in any direction.

Specimen cooling stages:
Airlock-exchangeable specimen cartridges can be cooled by liquid N_2 down to 150 K with double-tilting facilities up to 10°, or down to 120 K without tilting.

Transversal magnetic field stage:
On inserting this stage into a specially shielded objective pole piece, a horizontal magnetic field of up to 30 kA/m can be applied across the specimen plane and rotated mechanically into all azimuths.

Tensile stage:
A specimen can be fixed to a non-tiltable stage where a blade spring extends the specimen, controlled by heating a wire.

After some years of work and respective experience, a number of new stages were laboratory-made in order to meet particular requirements and to extend the facilities.

2.2.1. Heating/tilting stage

To attain higher temperatures, a new stage was built in 1983 [1] where the tantalum/ molybdenum specimen cartridge is heated by electron bombardment from a surrounding W filament, a method which was introduced into the design of *in situ* stages by Fujita and Komatsu [2]. The cartridge holder, including the filament heating and high-voltage leads as well as water cooling and a thermocouple, can be double-tilted up to 23° via the tongs mechanism shown in Fig. 2. The cartridge itself is exchangeable via the redesigned specimen airlock transfer mechanism. Temperatures of

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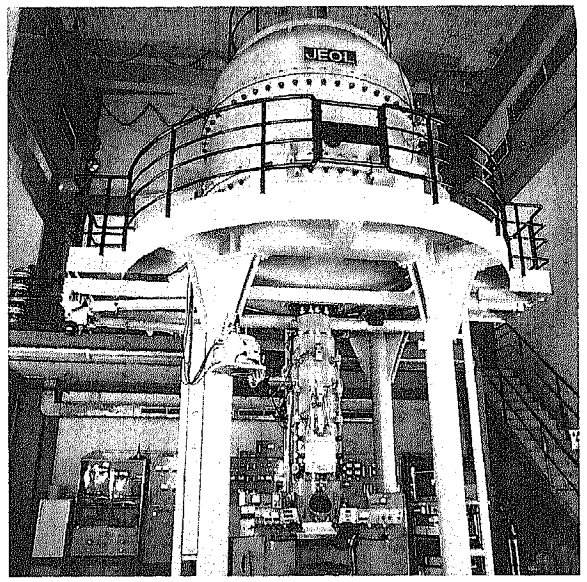

Fig.1. The JEOL 1000 kV high-voltage transmission electron microscope installed in Halle (Saale), Germany.

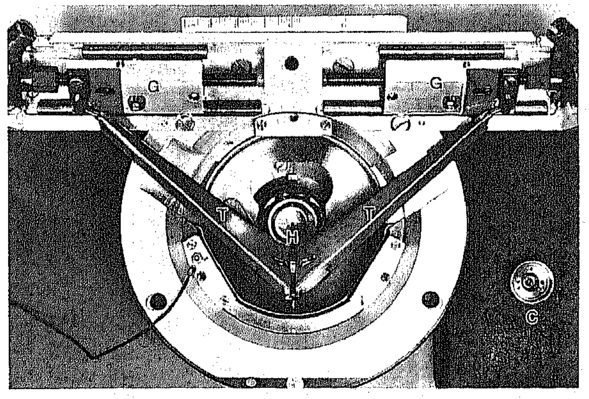

Fig.2. Laboratory-made universal 45° double-tilting stage viewed from the top. The specimen cartridge C shown separately is airlock-transferred to the cartridge holder H. It is tilted on moving the tongs T by means of gears G (G. Kästner, 1977).

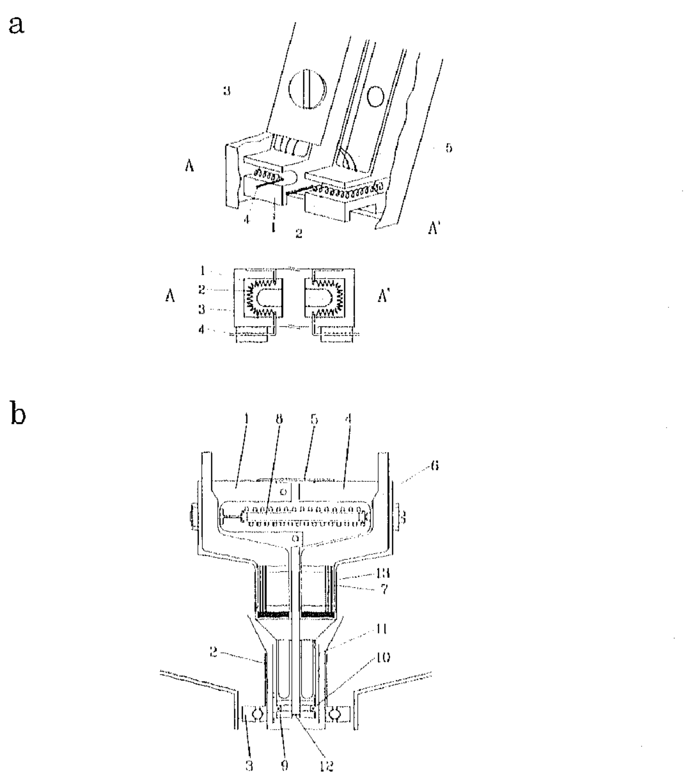

Fig.3. Schematic drawing of the double-tilting high-temperature straining stage. a) Perspective view of the hot zone. 1 - specimen grips, 2 - coil filaments, 3 - thermal shields, 4 - electric connections of filaments. b) Drive and cooling system. 1 - fixed lever, 2 - tilted cone carrying the deformation device, 3 - ring of cardanic suspension of the double-tilting stage, 4 - moveable lever, 5 - leaf spring, 6 - cooling water pipe, 7 - lamellae heat exchangers, 8 - stainless steel tube with heating coil, 9 - W-27Re specimen grips, 10 - W filaments, 11 - thermal shields, 12 - specimen, 13 - seminconducting strain gauges (U. Messerschmidt, 1994, after [4]).

1470 K were routinely applied.

2.2.2. Quantitative straining/tilting stage

A double-tilting straining stage was designed in 1975 [3]. Double-tilting of about 23° in most azimuth directions is possible owing to design features similar to those of the universal goniometer described above. The deformation is driven by the thermal expansion of an aluminium rod operating against a water cooling system. The displacement is transmitted to the specimen grips and enlarged by two symmetrical levers resulting in a relatively high elastic stiffness and a smooth action. Two full bridges of semiconducting strain gauges allow the measurement of load and specimen elongation. These features are sometimes necessary for interpreting the results, e.g., during unloading studies, but are always very helpful in performing the experiments.

2.2.3. High-temperature straining/tilting stage

To enable the study of the plastic deformation of high-temperature materials, including ceramics, a stage was designed for temperatures above 1300 K using again electron bombardment to heat the specimen grips [4]. **Fig. 3** shows an outline of this stage. Two small coil filaments (2 in Fig. 3a) are attached to the thermal shields (3). The electron current hits the specimen grips (1 in Fig. 3a, 9 in Fig. 3b). As for the room temperature stage described in the foregoing section, the deformation is driven by thermal expansion, this time of a stainless steel tube (8 in Fig. 3b). The load is transmitted to the specimen (12) by a fixed (1) and a movable lever (4). The heat of the thermal drive and of the hot specimen grips is carried off by water pipes (6) and small lamellae heat exchangers (7). The load is measured by a full bridge of semiconducting strain gauges (13). The whole stage is controlled via a personal computer. It has a maximum load of 15 N. Experiments have been performed at grip temperatures of up to 1520 K.

2.2.4. Electric fields

The 45° goniometer shown in Fig. 2 has been equipped with electrical contacts and with a special cartridge that allows one to apply a voltage of up to 1 kV to the specimen.

An attachment was built where the specimen is a fine metal tip which can be wetted with a liquid metal. On applying high voltage to the tip, one can observe the field-induced formation of a liquid metal cone. Image distortion by the electric field of the tip is reduced owing to the high acceleration voltage of the microscope.

2.2.5. Transverse bending stage

A bending stage was designed by Eckhard Langner of the Halle Branch of the Fraunhofer Institute of Mechanics of Materials in about 1993. The stage was mainly used to study the dislocation generation at cracks in silicon.

2.2.6. Environmental cell

To study the oxidation of metals, an environmental cell was designed for the HVEM [5]. To separate the gas near the specimen from the vacuum of the microscope, a differential pumping system with two inner and outer apertures each is applied using an additional turbo pump. The specimen is heated by a small furnace of alumina carrying a platinum heating coil. The system allows gas pressures between 10^{-3} and 150 Pa and temperatures above 1300 K.

The *in situ* techniques of the Halle HVEM are open for the cooperation with guest scientists from other institutions.

3. Scientific output

3.1. Further education

The facilities reported above as well as further instruments of the institute comprising scanning and high-resolution transmission electron microscopy soon became attractive to researchers not only of the GDR but of various formerly socialist countries. Consequently, an "International Centre of Electron Microscopy of the Socialist Countries" at the Institute of Solid State Physics and Electron Microscopy in Halle (Saale) was initiated in 1975 by Heinz Bethge, the director of the institute. The centre coordinated scientific collaboration and organized annual spring and autumn schools for training electron microscopists. The centre was headed by H. Bethge from 1975 to 1984 and by J. Heydenreich from 1984 to 1995. The centre was supervised by a Scientific Council of scientists from the member countries. This framework was also useful to overcome the stringent governmental restrictions on inviting scientists and lecturers from non-socialist countries. A total number of about 35 autumn or spring schools, resp., with 100 participants on an average, were held until 1995, including two International Symposia (in 1979, on *In situ* High Voltage Electron Microscopy in Materials Science, and in 1989, on Electron Microscopy in Plasticity and Fracture Research of Materials). In 1995, the centre was reorganized by forming an International Centre of Materials Science and Electron Microscopy, supported by the Max Planck Society and headed by Ulrich Gösele.

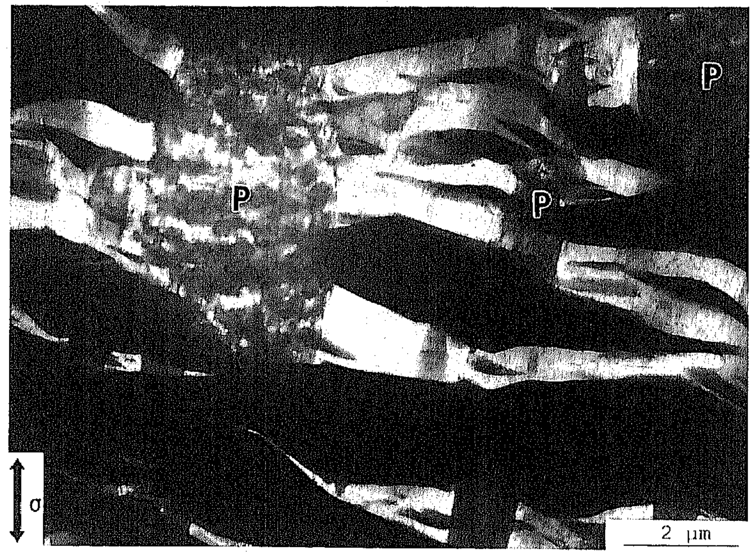

Fig.4. Thin section of high-impact polystyrene: Tensional strain (applied in vertical direction) is elongating the globular rubber particles P (of about 2 μm in size) and opening crazes (bright, horizontal layers). Within each craze, fine fibrillae represent polystyrene heavily elongated by strain. Without the optimized fraction of rubber particles, the crazes would elongate catastrophically until the material breaks (G. Michler, 1976).

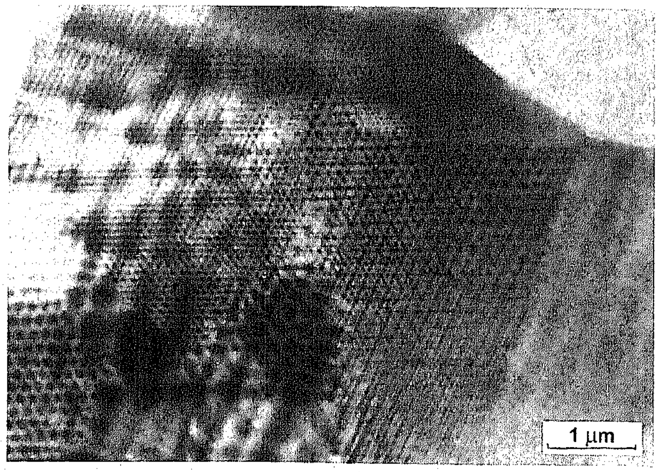

Fig.5. Liquid-crystal thin film prepared by vapour deposition onto a carbon film. The rod-shaped organic molecules of $C_7H_{15}O\text{-}C_6H_4\text{-}CH{=}N\text{-}(C_6H_4)_2\text{-}CN$ are hexagonally packed within smectic layers of finite grain size. Overlapping layers give rise to Moiré patterns. Accelerating voltage 1000 kV (G. Kästner, Ch. Dietzsch, 1980).

3.2. Methodical work

Since the HVEM is chiefly applied to study crystal defects by means of diffraction contrast, the operating contrast mechanisms had to be studied both experimentally and theoretically with particular emphasis on many-beam diffraction, on the anomalous absorption of thick specimens, and on kinematical bright-field imaging — a convenient technique similar but complementary to weak-beam dark-field imaging. A comprehensive theoretical treatment as well as experimental illustrations and tabulated many-beam extinction distances for electron energies of up to 1000 keV are given in [6].

3.3. Routine applications of the HVEM

Because of the activity of the "International Centre of Electron Microscopy", the HVEM facilities in Halle were easily accessible to many scientists from the socialist countries for almost 20 years. Thus, the applications covered a wide field of materials research including semiconductors, metals, ceramics, polymers, and thin films. It would be useless here to refer to the great variety of routine investigations. Instead, two examples of non-standard applications will be given.

Elevated acceleration voltages (electron energies) reduce the ionizing electron damage to organic specimens. Therefore, an HVEM is useful to transmit such specimens at a reduced damage rate or for a longer time. For this reason, since about 1975 Georg Michler and coworkers (Martin Luther University of Halle (Saale)) [7] performed *in situ* straining experiments on various polymers, using the stage supplied by the manufacturer. **Figure 4** shows an example of these studies.

For the same reason of reduced electron ionization damage, liquid crystals of the smectic type (of low vapour pressure) have been transmitted for the first time [8]. Certain types of dislocations were identified by means of diffraction contrast experiments, and the crystallographic space groups were determined from selected area electron diffraction patterns. An example is given in **Fig. 5**.

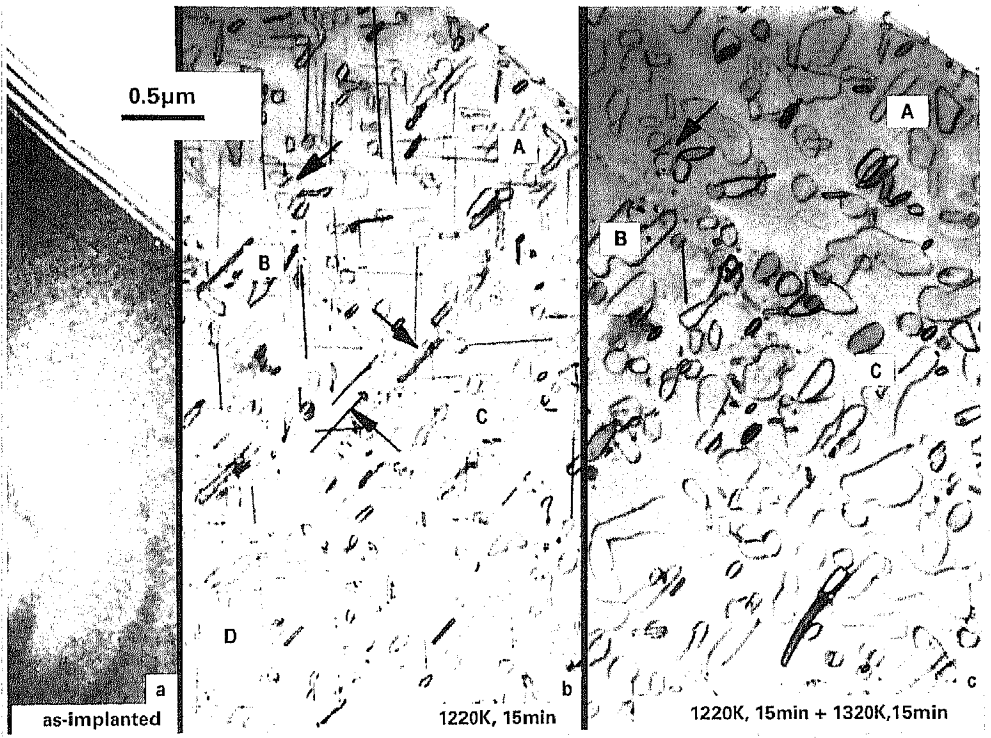

Fig.6. *In situ* annealing of Si implanted with 10^{15} B^+/cm^2 at 150 keV. Letters designate equal places in b) and c) (D. Hoehl, 1989).

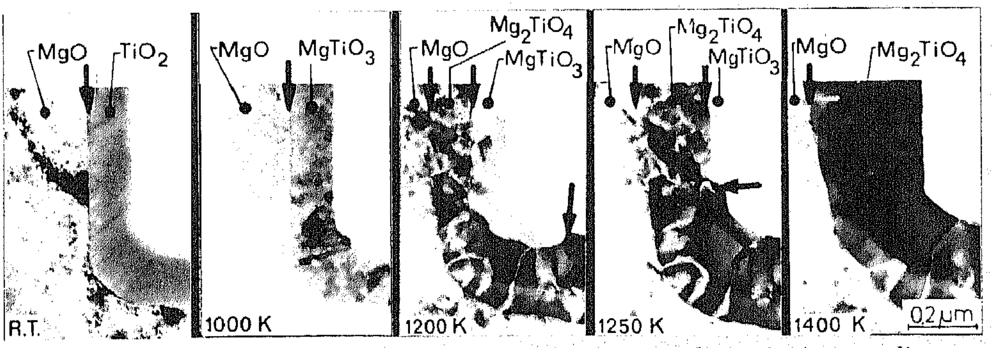

Fig.7. Solid state reactions in an MgO/TiO_2 cross section specimen during *in situ* annealing (D. Hesse, L. Berthold, D. Hoehl, 1994).

3.4. Applications of laboratory-made *in situ* devices

The stages described in Section 2.2 have largely been used to study different topics and materials. Only a few can be mentioned here.

3.4.1. Annealing studies

The high-temperature heating/tilting stage (cf. 2.2.1.) has been used to study various annealing problems, e.g., the recrystallization of ion-implanted silicon. Depending on the implantation conditions (i. e., the ion dose, energy and mass) the crystalline material is damaged (up to amorphization) so that it has to be annealed. **Figure 6** shows the interaction and transformation of dislocations that are created during *in situ* annealing of ion-implanted silicon in an identical specimen area [9]. The antiparallel arrows indicate the transformation of a rod-like defect into an elongated dislocation dipole. Dislocation loops can glide to the surface, dislocation segments of opposite sign interact and annihilate, leading to the intended reduction of the dislocation density.

Solid state reactions were observed in cross section specimens of a TiO_2 layer on an MgO substrate as shown in **Fig. 7**. The interface is sharp at room temperature (R.T.). At about 1000 K a polycrystalline layer of $MgTiO_3$ forms rapidly followed by the slow growth of a single crystal of the stable phase of Mg_2TiO_4 at higher temperatures [10]. Such processes at temperatures above 1300 K cannot be studied by conventional *in situ* heating stages.

3.4.2. Deformation at room temperature

Very detailed studies were performed on the plastic deformation of magnesium oxide single crystals. As shown in the sequence of images of **Fig. 8**, dislocations bow out between obstacles under load. Due to the cusps, forming at the obstacles, forces are exerted on them. After their thermally activated overcoming, new equilibrium positions are occupied. Detailed quantitative data were obtained from such micrographs and from video recordings, and compared with current theories. These data include the statistical frequency distributions of the distances between the obstacles and the forces acting on them, the distribution of jump distances during motion, the orientation dependence of the line tension of dislocations [11], or radiation-hardening inside the HVEM.

Other materials studied comprise Ni, age-hardened Al-Mg-Zn and Al-Ag alloys and Al-Li alloys with precipitates of the ordered phase Al_3Li. The results of the *in situ* experiments yielded quantitative data on the interaction between dislocations and precipitates. In some cases, they showed that the deformation is controlled by other defects than those considered before the results of the *in situ* experiments were known.

3.4.3. Deformation at high temperatures

In situ straining tests at high temperatures were performed on a number of materials including TiAl materials of different microstructures, NiAl, NiAl containing Ta, an oxide dispersion strengthened material, ZrO_2-Y_2O_3 alloys of different microstructures, and Al-Pd-Mn single quasicrystals.

ZrO_2-Y_2O_3 crystals may contain precipitates of tetragonal crystal structure which form a complicated domain structure. In the so-called t'-ZrO_2, this domain structure fills the whole crystal volume. During loading at high temperatures, the tetragonal domains may switch the orientations of their c-axes, giving rise to ferroelastic deformation. **Fig. 9**, taken during *in situ* deformation at 1420 K, shows the original domain structure in strong contrast in the lower part of the figure. The transformed regions form a tetragonal single crystal so that the strong electron microscopy contrast disappears. Details of this ferroelastic deformation are described in [12]. The ferroelastic deformation was proven for the first time also in the tetragonal precipitates in partially stabilized zirconia. It changes the microstructure for the subsequent dislocation mechanisms so that the models of precipitation hardening in these materials have to be revised.

At present, a challenging problem is the understanding of the microprocesses of the plastic deformation of quasicrystals. According to the non-periodic structure of these materials, dislocations have to produce a special kind of structural disorder during their motion. Therefore, they have been considered immobile. *In situ* straining experiments at high temperatures gave the first direct evidence that, nevertheless, the dislocations carry the deformation also in quasicrystals [13]. Further details of the dislocation motion are being studied.

In conclusion, the described *in situ* experiments in the HVEM gave important insight into the deformation processes, frequently also by observing very different mechanisms in different temperature ranges.

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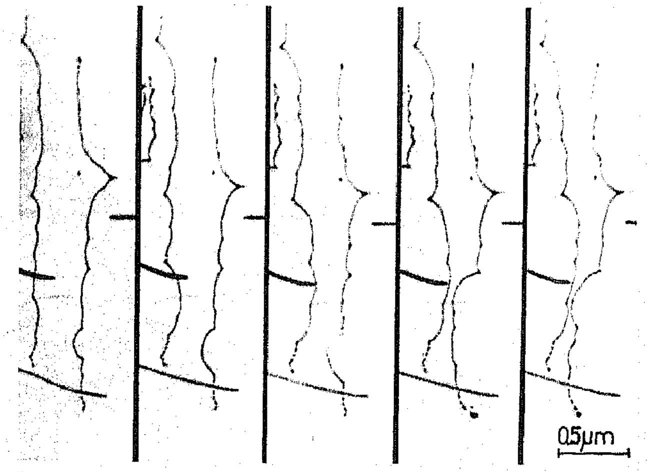

Fig.8. Moving dislocations in a magnesium oxide single crystal during deformation inside the HVEM at room temperature. The dislocations bow out between small precipitates where they form cusps. The dislocations move forward by overcoming the obstacles individually and assuming new equilibrium positions (U. Messerschmidt, F. Appel, 1976).

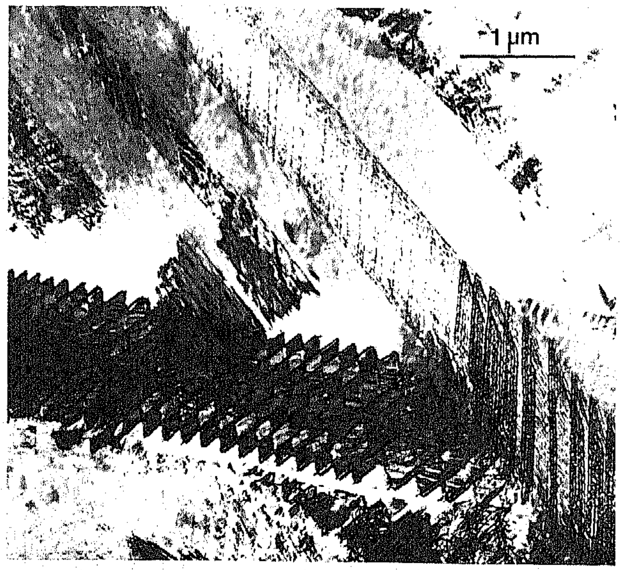

Fig.9. Ferroelastic deformation of t' zirconia during *in situ* straining in the HVEM at 1420 K. The dark structures are the untransformed tetragonal domains. Bright areas are transformed by ferroelastic deformation, which reduces the electron microscopy diffraction contrast (D. Baither, B. Baufeld, M. Bartsch, U. Messerschmidt, 1995).