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Ludwig Reimer

Transmission Electron Microscopy

Physics of Image Formation and Microanalysis

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With 264 Figures

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Preface

The aim of this book is to outline the physics of image formation, electron-specimen interactions and image interpretation in transmission electron microscopy. The book evolved from lectures delivered at the University of Münster and is a revised version of the first part of my earlier book *Elektronenmikroskopische Untersuchungs- und Präparationsmethoden*, omitting the part which describes specimen-preparation methods.

In the introductory chapter, the different types of electron microscope are compared, the various electron-specimen interactions and their applications are summarized and the most important aspects of high-resolution, analytical and high-voltage electron microscopy are discussed.

The optics of electron lenses is discussed in Chapter 2 in order to bring out electron-lens properties that are important for an understanding of the function of an electron microscope. In Chapter 3, the wave optics of electrons and the phase shifts by electrostatic and magnetic fields are introduced; Fresnel electron diffraction is treated using Huygens' principle. The recognition that the Fraunhofer-diffraction pattern is the Fourier transform of the wave amplitude behind a specimen is important because the influence of the imaging process on the contrast transfer of spatial frequencies can be described by introducing phase shifts and envelopes in the Fourier plane. In Chapter 4, the elements of an electron-optical column are described: the electron gun, the condenser and the imaging system.

A thorough understanding of electron-specimen interactions is essential to explain image contrast. Chapter 5 contains the most important facts about elastic and inelastic scattering and x-ray production. The origin of scattering and phase contrast of non-crystalline specimens is described in Chapter 6. High-resolution image formation using phase contrast may need to be completed by image-reconstruction methods in which the influence of partial spatial and temporal coherence is considered.

Chapter 7 introduces the most important laws about crystals and reciprocal lattices. The kinematical and dynamical theories of electron diffraction are then developed. Electron diffraction is the source of diffraction contrast, which is important for the imaging of lattice structure and defects and is treated in Chapter 8. Extensions of the capabilities of the instrument have awakened great interest in analytical electron microscopy: x-ray microanalysis, electron-energy-loss spectroscopy and electron diffraction, summarized in Chapter 9. The final Chapter 10 contains a brief account of the various specimen-damage processes caused by electron irradiation.

Electron microscopy is an interdisciplinary science with a strong physical background. The full use of all its resources and the interpretation of the results requires familiarity with many branches of knowledge. Physicists are in a favoured situation because they are trained to reduce complex observations to simpler models and to use mathematics for formulating "theories". There is thus a need for a book that expresses the contents of these theories in language accessible to the "normal" electron microscope user. However, so widespread is the use of electron microscopy that there is no such person as a normal user. Biologists will need only a simplified account of the theory of scattering and phase contrast and of analytical methods, whereas electron diffraction and diffraction contrast used by material scientists lose some of their power if they are not presented on a higher mathematical level. Some articles in recent series on electron microscopy have tried to bridge this gap by over-simplification but this can cause misunderstandings of just the kind that the authors wished to avoid. In the face of this dilemma, the author decided to write a book in his own physical language with the hope that it will be a guide to a deeper understanding of the physical background of electron microscopy.

A monograph by a single author has the advantage that technical terms are used consistently and that cross-referencing is straightforward. This is rarely the case in books consisting of review articles written by different specialists. Conversely, the author of a monograph is likely to concentrate, perhaps unconsciously, on some topics at the expense of others but this also occurs in multi-author works and reviews. Not every problem can be treated on a limited number of pages and the art of writing such a book consists of selecting topics to omit. I apologize in advance to any readers whose favourite subjects are not treated in sufficient detail. The number of electron micrographs has been kept to a minimum; the numerous simple line drawings seem better suited to the more theoretical approach adopted here.

There is a tendency for transmission electron microscopy and scanning electron microscopy to diverge, despite their common physical background. Electron microscopy is not divided into these categories in the present book but because transmission and scanning electron microscopy together would increase its size unreasonably, only the physics of the transmission electron microscope is considered – the physics of its scanning counterpart will be examined in a complementary volume.

A special acknowledgement is due to P. W. Hawkes for his cooperation in revising the English text and for many helpful comments. Special thanks go to K. Brinkmann and Mrs. R. Dingerdissen for preparing the figures and to all colleagues who gave me permission to publish their results.

Münster, July 1982 *L. Reimer*

Contents

1. Introduction

1.1 Types of Electron Microscopes

Although the main concern of this book is transmission electron microscopy, the functions and limits of the other types of electron microscopes are also mentioned in this introductory chapter to show the advantages and disadvantages of their various imaging techniques. Several types of electron microscopes and analysing instruments capable of furnishing an "image" can be distinguished. We now examine these briefly, in turn, without considering the historical sequence in which these instruments were developed. In these background sections, references are restricted to review articles and books.

1.1.1 Electron Microscopes for the Direct Imaging of Surfaces of Bulk Specimens

a) Emission Electron Microscopes [1.1, 2, 10]

In an emission electron microscope (Fig. 1.1 a), the cathode that emits the electrons is directly imaged by an electrostatic immersion lens, which accelerates the electrons and produces an intermediate image of the emission intensity distribution at the cathode. This image can be magnified by further electron lenses and is observed on a fluorescent screen or with an image intensifier. The cathode (specimen) has to be plane and its surface should not be too irregular. The electron emission can be stimulated by

a) heating the cathode (thermionic emission), which means that observation is possible only at elevated temperatures and for a limited number of materials; alternatively, the electron-emission temperature need not be raised beyond 500–1000 °C if a thin layer of barium is evaporated on the surface because this lowers the work function;

b) secondary-electron excitation by particle bombardment, by irradiating the cathode surface with an additional high-energy electron beam or an ion beam at grazing incidence;

c) irradiation of the cathode with an ultra-violet light source to excite photoelectrons (photoelectron-emission microscope PhEEM [1.3, 4])

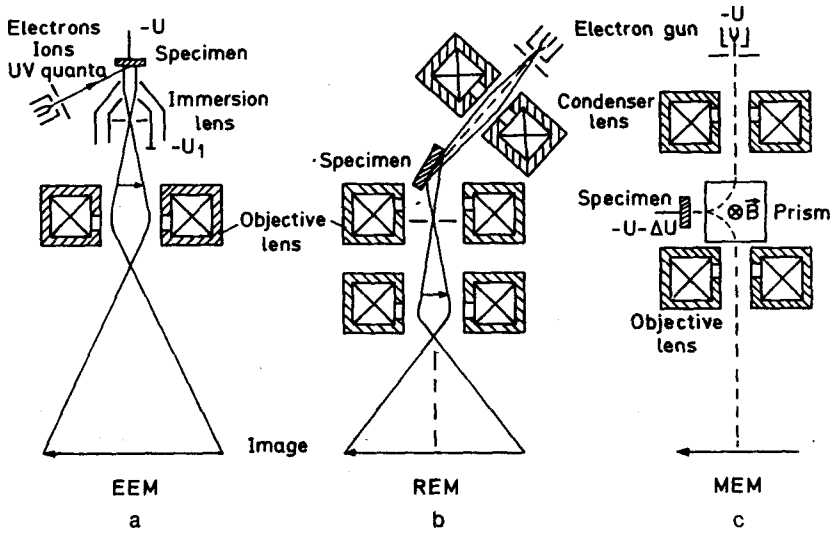

Fig. 1.1 a–c. Schematic ray paths for (**a**) an emission electron microscope (EEM), (**b**) a reflexion electron microscope (REM), (**c**) a mirror electron microscope (MEM)

These instruments have a number of interesting applications but their use is limited to particular specimens; at present, therefore, scanning electron microscopes (Sect. 1.1.2) are the most widely used instruments for imaging bulk specimens, especially because there is no need to limit the roughness of the specimen surface. The final restriction is the limited number of electrons emitted, which restricts the image intensity at high magnification, and the resolution of the immersion lens system is only of the order of 10–30 nm. On the credit side, surfaces can be observed directly in-situ and each of the processes a–c) generates a specific contrast. The photoelectron-emission electron microscope has the advantage of being applicable to nearly any flat specimen surface, including biological specimens [1.5, 6]. The image contrast is caused by differences of the emission intensity (material and crystal orientation contrast) and by angular selection with a diaphragm that intercepts electrons whose trajectories have been deflected by variations of the equipotentials near the surface caused by surface steps (topographic contrast), surface potentials (potential contrast) or magnetic stray fields (magnetic contrast).

b) Reflexion Electron Microscopes [1.7–10]

The electrons that emerge as a result of primary-electron bombardment are either low-energy secondary electrons, which can be imaged in an emission electron microscope (see above) or scanning electron microscope (see below), or primary (backscattered) electrons with large energy losses, which cannot be focused sharply by an electron lens because of chromatic aberra-

tion. However, imaging of the surface is possible for grazing electron incidence below 10°, the "reflected" electrons being imaged with an objective lens (Fig. 1.1 b). The energy-loss spectrum of the reflected electrons has a half-width of the order of 100–200 eV. With additional energy selection by means of an electrostatic filter lens, a resolution of 10–20 nm can be attained [1.11]. Because the angle of incidence is so low, small surface steps can be imaged with high contrast. The angular distribution of the reflected electrons at single crystals is a reflexion high-energy electron diffraction (RHEED) pattern with Bragg diffraction spots; images exhibiting crystallographic contrast can be found by selecting individual Bragg spots. A transmission electron microscope can be operated in this mode by tilting the electron gun and condenser-lens system.

c) Mirror Electron Microscopes [1.12, 13]

An electron beam is deflected by a magnetic sector field, and retarded and reflected at a flat specimen surface, which is biased a few volts more negative than the cathode of the electron gun (Fig. 1.1 c). The reflected-electron trajectories are influenced by irregularities of the equipotential surfaces in front of the specimen, which can be caused by surface roughness or by potential differences and specimen charges; magnetic stray fields likewise act on the electron trajectories. An advantage of this method is that the electrons do not strike the specimen; it is the only technique that permits surface charges to be imaged undisturbed. After passing through the magnetic sector field again, the electrons can be selected according to their angular deflection. The lateral resolution of a mirror electron microscope is of the order of 50–100 nm. Single surface steps, 5 nm in height, can produce discernible contrast. Such a mirror electron microscope can be combined with an electron interferometer (Sect. 3.1.4), which offers the possibility of measuring phase shifts caused by the equipotential surfaces or magnetic stray fields with high precision. Methods of scanning mirror electron microscopy [1.14, 15] allow a more quantitative separation of the observed image point and the local beam deflection.

1.1.2 Instruments Using Electron Microprobes

a) Scanning Electron Microscopes (SEM) [1.16–31]

SEM is the most important electron-optical instrument for the investigation of bulk specimens. An electron probe is produced by an one-, two- or three-stage demagnification of the smallest cross-section of the electron beam after acceleration. This electron probe, 5–10 nm in diameter if a thermionic electron gun is used and 0.5–2 nm with a field-emission gun, is scanned in a raster over a region of the specimen (Fig. 1.2). The smallest diameter of the electron probe is limited by the minimum acceptable electron probe current of

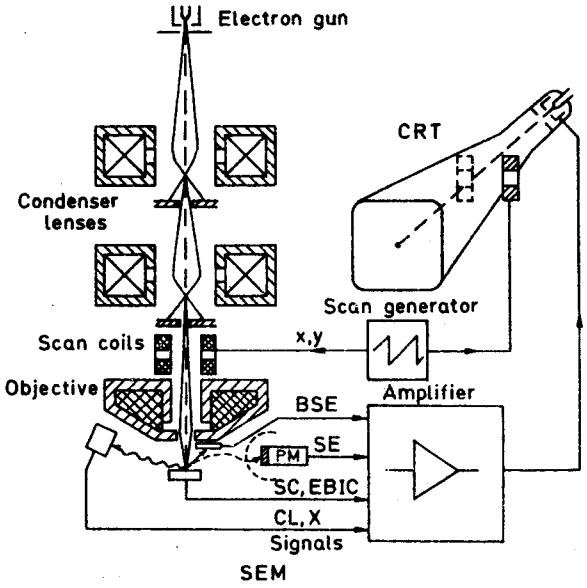

Fig. 1.2. Schematic ray path for a scanning electron microscope (SEM)

10^{-12}–10^{-11}A. It is determined by the need to generate an adequate signal-to-noise ratio, and by the spherical aberration of the final probe-forming lens. The image is displayed on a cathode-ray tube (CRT) rastered in synchronism. The CRT beam intensity can be modulated by any of the different signals that result from the electron-specimen interactions.

The most important signals are produced by secondary electrons with most probable exit energies of 2–5 eV and by backscattered electrons, with energies that range from the energy of the primary electrons to about 50 eV. The secondary-electron yield and the backscattering coefficient depend on the angle of electron incidence (topographic contrast), the crystal orientation (channelling contrast) and electrostatic and magnetic fields near the surface (voltage and magnetic contrast). A signal can also be produced by the specimen current and by electron-beam-induced currents in semiconductors. Analytical information is available from the x-ray spectrum and Auger electrons or from light quanta emitted by cathodoluminescence. The crystallographic structure and orientation can be obtained from electron channelling patterns or electron back-scattering patterns (Sect. 9.3.4) and from x-ray Kossel diagrams.

The resolution of the different modes of operation and types of contrast depends on the information volume that contributes to the signal. Secondary electrons provide the best resolution, because the exit depth is very small, of the order of a few nanometres. The exit volume limits the resolution of field-emission systems with electron-probe diameters smaller than 2 nm. The information depth of backscattered electrons is much greater, of the order of half the electron range, which is as much as 0.1–1 μm, depending on the

density of the specimen and the electron energy. The secondary-electron signal also contains a large contribution from the backscattered electrons when these penetrate the surface layer. This is why SEMs mostly operate in the range $E = 10 - 20$ keV. At higher energies, the electron range and the diameter of the electron-diffusion region are greater. Conversely, higher energies are of interest for x-ray microanalysis if the K shells of heavy elements are to be excited. Decreasing the electron energy has the advantage that information can be extracted from a volume nearer to the surface, but the diameter of the electron probe increases owing to the decrease of gun brightness.

Unlike transmission electron microscopy, special specimen-preparation techniques are rarely needed in scanning electron microscopy. Nevertheless, charging effects have to be avoided, by coating the specimen with a thin conductive film for example and organic specimens have to be protected from surface distortions by fixation or cryo-techniques.

b) X-ray and Auger-Electron Microanalysers [1.23–28]

By using a wavelength-dispersive x-ray spectrometer (Bragg reflection at a crystal), we can work with high x-ray excitation rates and electron-probe currents of the order of 10^{-8}–10^{-7} A, though the electron-probe diameter is then larger, about 0.1–1 μm. The main task of an x-ray microanalyser is to analyse the elemental compositions of flat, polished surfaces at normal electron incidence with a high analytical sensitivity. The ray diagram of such an instrument is similar to that of a SEM but two or three crystal spectrometers, which can simultaneously record different characteristic x-ray wavelengths, are attached to the column. The surface can be imaged by one of the SEM modes to select the specimen points to be analysed.

SEM or x-ray microanalyser can be equipped with an Auger-electron spectrometer, of the cylindrical mirror type for example. It is then necessary to work with ultra-high vacuum in the specimen chamber because Auger electrons are extremely sensitive to the state of the surface: a few atomic layers are sufficient to halt them. Special Auger-electron microanalysers have therefore been developed, in which the 1–10 keV electron gun may for example be incorporated in the inner cylinder of a spectrometer. This type of instrument can also work in the scanning mode so that an image of the surface can be formed with secondary electrons or an element-distribution map can be generated using Auger electrons.

1.1.3 Transmission Electron Microscopes

a) Conventional Transmission Electron Microscope [1.32–96]

In a conventional transmission electron microscope (CTEM, or TEM for short) (Figs. 1.3 and 4.17c, 20), a thin specimen is irradiated with an electron

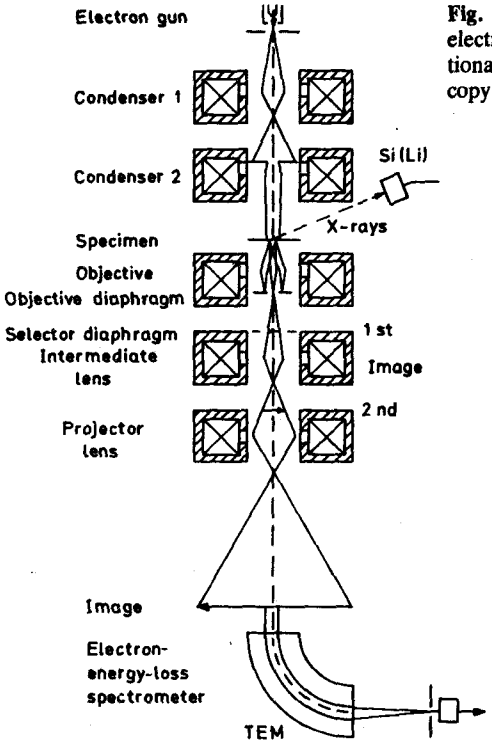

Fig. 1.3. Schematic ray path for a transmission electron microscope (TEM) equipped for additional x-ray and electron energy-loss spectroscopy

beam of uniform current density; the electron energy is in the range 60–150 keV (usually 100 keV) or 200 keV–3 MeV in the case of the high-voltage electron microscope (HVEM).

Electrons are emitted in the electron gun (Sect. 4.1) by thermionic emission from tungsten hairpin cathodes or LaB_6 rods or by field emission from pointed tungsten filaments. The latter are used when high gun brightness is needed. A two-stage condenser-lens system permits variation of the illumination aperture and the area of the specimen illuminated (Sec. 4.2). The electron-intensity distribution behind the specimen is imaged with a three- or four-stage lens system, onto a fluorescent screen (Sect. 4.4). The image can be recorded by direct exposure of a photographic emulsion inside the vacuum (Sect. 4.6).

The lens aberrations of the objective lens are so great that it is necessary to work with very small objective apertures, of the order of 10–25 mrad, to achieve resolution of the order of 0.2–0.5 nm. Bright-field contrast is produced either by absorption of the electrons scattered through angles larger than the objective aperture (scattering contrast) or by interference between the scattered wave and the incident wave at the image point (phase contrast). The phase of the electron waves behind the specimen is modified by the wave