

**Advances in Optical and  
Electron Microscopy**

**Volume 12**

*Advances in*

**OPTICAL *and* ELECTRON  
MICROSCOPY**

This page intentionally left blank

*Advances in*  
**OPTICAL *and* ELECTRON  
MICROSCOPY**

*Volume 12*

*Edited by*

**T. MULVEY**

*Aston University, Birmingham, UK*

AND

**C. J. R. SHEPPARD**

*The University of Sydney, Sydney, Australia*



**ACADEMIC PRESS**

*Harcourt Brace Jovanovich, Publishers*

London · San Diego · New York · Boston

Sydney · Tokyo · Toronto

ACADEMIC PRESS LIMITED  
24/28 Oval Road,  
LONDON NW1 7DX

*U.S. Edition Published by*  
ACADEMIC PRESS INC.  
San Diego, CA 92101

Copyright © 1991 by ACADEMIC PRESS LIMITED

This book is printed on acid-free paper

*All Rights Reserved*

No part of this book may be reproduced or transmitted in any form or by any means, electronic or mechanical, including photocopy, recording, or any information storage and retrieval system without permission in writing from the publisher

**British Library Cataloguing in Publication Data**

Advances in optical and electron microscopy.  
—Vol. 12  
1. Microscopy  
502.8

ISBN 0-12-029912-7  
ISSN 0065-3012

Typeset by P&R Typesetters Ltd, Salisbury, Wilts  
Printed by Galliard (Printers) Ltd, Great Yarmouth, Norfolk

## Contributors

- L. DUBBELDAM, *Faculty of Applied Physics, Delft University of Technology, P.O. Box 5046, NL-2600 GA Delft, The Netherlands. ( Present address: Space Research Organization of the Netherlands, Niels Bohrweg 2, P.O. Box 9504, 2300 RA Leiden, The Netherlands. )*
- J. HARTIKAINEN, *Department of Physics, University of Helsinki, Siltavuorenpenger 20 D, SF-00170 Helsinki, Finland.*
- J. JAARINEN, *Department of Physics, University of Helsinki, Siltavuorenpenger 20 D, SF-00170 Helsinki, Finland.*
- P. KRUIT, *Department of Applied Physics, Delft University of Technology, Lorentzweg 1, NL-2628 CJ Delft, The Netherlands.*
- H. LICHTER, *Institut für Angewandte Physik, Universität Tübingen, Auf der Morgenstelle 12, D-7400 Tübingen, Federal Republic of Germany.*
- M. LUUKKALA, *Department of Physics, University of Helsinki, Siltavuorenpenger 20 D, SF-00170 Helsinki, Finland.*
- G. MÖLLENSTEDT, *Institut für Angewandte Physik, Universität Tübingen, Auf der Morgenstelle 12, D-7400 Tübingen, Federal Republic of Germany.*
- D. W. POHL, *IBM Research Division, Zurich Research Laboratory, CH-8803 Rüschlikon, Switzerland.*

This page intentionally left blank

## Preface

Progress and innovation in optical and electron microscopy at a fundamental level still manage to surprise even experts in the field. The present volume, for example, records a world first: the complete realization, at the University of Tübingen, of Gabor's original concept, announced just over 40 years ago, of correcting the spherical aberration of an electron microscope by means of electron beam holography and thereby achieving atomic resolution, in which both the phase and the amplitude of the wave scattered by the specimen are faithfully reproduced in the image. This is indeed a milestone in the development of the microscope and the interpretation of its image. Möllenstedt's seminal contribution to this method, his invention of the electron Fresnel biprism, is set out in an accompanying paper.

Progress in scanning electron microscopy is also reported. Recently, the ability to extract and display information contained in the secondary electron emission from the specimen in an SEM or STEM has also improved. This has been greatly helped by the possibility of using a "moving axis" probe-forming lens, by means of which the effective axis of the lens may be deflected laterally by suitably placed deflecting coils; this reduces or eliminates the usual off-axis aberrations. The underlying theory and some applications are set out in the review by P. Kruit; this is complemented by L. Dubbeldam's review in which a new method for non-contact voltage measurement of integrated circuits in the SEM is highlighted. Many people believe that the light microscope is inherently limited in resolution to about a quarter of a wavelength of light. One of the most fruitful recent developments is the scanning near-field optical microscope (SNOM) in which a specimen is raster scanned in the near vicinity—within 20 nm or so—of a point source. Such a microscope, capable of a resolution of 20–50 nm is described by D. W. Pohl. The SNOM could prove very useful in connection with the development of nano-engineering structures that are now being investigated in many countries.

Finally, J. Hartikainen and colleagues address the question of non-destructive testing by thermal wave microscopy. In this technique a modulated heating produced using laser illumination results in temperature variations which propagate through the specimen as thermal waves, thus giving spatial information concerning its thermal and acoustic properties. This is one of several modern techniques of microscopy, neither optical nor electron, which can greatly increase our understanding of a specimen's structure.

New forms of microscopy, or the practical realization of older forms that were not technologically feasible in the past, are a feature of the present era; the editors are committed to a policy of including reviews in this area.

T. MULVEY  
C. J. R. SHEPPARD

# Contents

CONTRIBUTORS . . . . .	v
PREFACE . . . . .	vii

## The Invention of the Electron Fresnel Interference Biprism

G. MÖLLENSTEDT

I. Childhood years in Bielefeld . . . . .	1
II. Gaining technical know-how as a student apprentice . . . . .	2
III. A broad education in physics with Professor Walter Kossel . . . . .	2
IV. Influence of the work of Kikuchi and convergent beam diffraction . . . . .	3
V. Electron-optical experiments with Brüche, Scherzer and Mahl . . . . .	5
VI. Quantitative testing of the operation of the biprism . . . . .	12
VII. Measurement of the inner potential of solids . . . . .	19
VIII. Electron interference microscope in the transmission mode . . . . .	21
IX. The intensity problem in electron interferometers . . . . .	21
X. Atomic resolution electron holography . . . . .	21

## Electron Image Plane Off-axis Holography of Atomic Structures

HANNES LICHTÉ

I. Introduction . . . . .	25
II. Principles of off-axis image plane electron holography . . . . .	33
A. Taking the electron hologram . . . . .	34
B. Reconstruction of the electron image wave . . . . .	35
III. Performance of image plane electron holography . . . . .	41
A. Effect of restricted coherence . . . . .	42
B. Quantum noise . . . . .	47
C. Information transfer capacity . . . . .	47
D. Lateral resolution and field of view . . . . .	49
E. Artefacts in the recorded wave . . . . .	52
F. Problems of recording an electron hologram . . . . .	54
IV. Influence of the lens aberrations in the high-resolution domain . . . . .	60
A. Coherent aberrations . . . . .	61
B. Incoherent aberrations . . . . .	67
V. Reconstruction of the image wave and correction of aberrations . . . . .	69

VI.	Experimental realization of holography of atomic structures . . . . .	71
	A. Experimental set-up . . . . .	71
	B. Experimental results . . . . .	74
	C. First results with atomic structures of weak objects . . . . .	74
	D. Holographic imaging of strong objects . . . . .	78
	E. Numerical reconstruction, including a preliminary correction of aberrations . . . . .	78
VII.	Conclusion . . . . .	84
VIII.	List of symbols . . . . .	87
	References . . . . .	90

## **Magnetic Through-the-lens Detection in Electron Microscopy and Spectroscopy, Part 1**

P. KRUIT

I.	Introduction . . . . .	93
II.	Historical development of through-the-lens detection . . . . .	96
	A. High-efficiency detection of secondary electrons . . . . .	96
	B. Quantitative voltage-contrast measurements . . . . .	104
	C. Spectroscopy of emitted electrons . . . . .	107
III.	Historical development of the magnetic parallelizer for spectroscopy applications . . . . .	113
IV.	Theory of adiabatic motion . . . . .	119
	A. Introduction . . . . .	119
	B. Simple perturbation approach . . . . .	121
	C. Northrop's perturbation method . . . . .	128
	D. General theory of adiabatic invariants . . . . .	129
	E. Conclusions from adiabatic theory . . . . .	130
V.	Summary . . . . .	134
	References . . . . .	135

## **Advances in Voltage-contrast Detectors in Scanning Electron Microscopes**

LUC DUBBELDAM

I.	Introduction . . . . .	140
II.	Test techniques for integrated circuits . . . . .	140
	A. Overview . . . . .	140
	B. Techniques with secondary electrons . . . . .	142

	C. Electron-optical measurements . . . . .	149
	D. Electron and optical beam-induced current . . . . .	150
	E. Reconfiguration and mask repair . . . . .	152
	F. Metrology . . . . .	154
III.	Secondary electrons . . . . .	155
	A. Secondary electron emission . . . . .	155
	B. Voltage-contrast detection . . . . .	165
	C. Time resolution . . . . .	181
IV.	Voltage-contrast detectors . . . . .	182
V.	Design of a double channel spectrometer . . . . .	191
	A. General design considerations . . . . .	191
	B. The column . . . . .	194
	C. The primary system . . . . .	195
	D. The magnetic parallelizer . . . . .	201
	E. The spectrometer . . . . .	206
VI.	Measurements . . . . .	216
	A. General measurement set-up . . . . .	216
	B. The scanning system . . . . .	219
	C. S-curves on both detectors . . . . .	219
	D. SE-spectrum on the lower detector . . . . .	222
	E. Voltage contrast on both detectors . . . . .	222
	F. Closed feedback loop . . . . .	224
	G. Signal/noise ratio for voltage measurements . . . . .	227
	H. Voltage-contrast isolation . . . . .	230
VII.	Future developments . . . . .	234
VIII.	Conclusions . . . . .	235
IX.	List of symbols . . . . .	237
	Acknowledgements . . . . .	237
	References . . . . .	238

## Scanning Near-field Optical Microscopy (SNOM)

D. W. POHL

I.	Introduction . . . . .	243
	A. Microscopy environment . . . . .	243
	B. SNOM principle and general properties . . . . .	246
	C. Potential areas of application . . . . .	247
	D. Organization of this paper . . . . .	247
II.	Historical background . . . . .	247
	A. SNOM proper . . . . .	248
	B. Antennas in front of a second medium . . . . .	249
	C. Elastic scattering of light . . . . .	250
	D. Transmission of small apertures . . . . .	250

	E. Scanning tunnelling microscopy and related techniques	250
	F. Evanescent wave microscopy	250
III.	Theoretical background	252
	A. General remarks	252
	B. Near field of an ideal dipole	252
	C. Dipole in front of a dielectric or conducting halfspace	254
	D. Small metallic particle in front of a dielectric or conducting halfspace	261
	E. Small apertures	264
	F. Pointed tips as NF optical probes	269
	G. Summary of theory	272
IV.	Experimental work	272
	A. Types of SNO microscope	272
	B. Basic NF optical experiments	283
	C. Plasmons and spectroscopic effects	287
	D. Imaging by SNOM	292
V.	Discussion	306
	A. State of the art	306
	B. Outlook	308
	Acknowledgements	309
	References	309

## Microscopic Thermal Wave Non-destructive Testing

JARI HARTIKAINEN, JUSSI JAARINEN AND MAURI LUUKKALA

I.	Introduction	313
II.	Thermal waves and their generation	315
III.	The resolution of the thermal wave microscope	321
IV.	Photothermal NDE techniques with periodic heating	325
	A. General	325
	B. Photoacoustic cell	325
	C. Photothermal radiometry	327
	D. Surface displacement techniques	329
	E. Piezoelectric detection	332
	F. Optical beam deflection	332
	G. Reflectance measurements	335
	H. Other techniques	338
V.	Photothermal pulse and scanning methods	338
	A. General	338
	B. Examples	340
	C. Laser scanning techniques	342
	D. Numerical methods	343
VI.	Experimental pulse techniques	345
	A. Traditional pulse techniques	345
	B. Flash techniques with an IR camera	347
	C. IR scanning techniques	351
VII.	Conclusion	352
	References	356
	INDEX	361

# The Invention of the Electron Fresnel Interference Biprism\*

G. MÖLLENSTEDT

*Institut für Angewandte Physik, Universität Tübingen, Auf der Morgenstelle 12, D-7400 Tübingen, Federal Republic of Germany*

I. Childhood years in Bielefeld . . . . .	1
II. Gaining technical know-how as a student apprentice . . . . .	2
III. A broad education in physics with Professor Walter Kossel . . . . .	2
IV. Influence of the work of Kikuchi and convergent beam diffraction . . . . .	3
V. Electron-optical experiments with Brüche, Scherzer and Mahl . . . . .	5
VI. Quantitative testing of the operation of the biprism . . . . .	12
VII. Measurement of the inner potential of solids . . . . .	19
VIII. Electron interference microscope in the transmission mode . . . . .	21
IX. The intensity problem in electron interferometers . . . . .	21
X. Atomic resolution electron holography . . . . .	21

## I. CHILDHOOD YEARS IN BIELEFELD

In order to describe how I happened on the idea of realizing interference with electron waves by means of an electron optical biprism, I must, by way of introduction, say a few words about my early physics education. I should begin with the remark that I had the good fortune to matriculate in the Oberrealschule Helmholtz at Bielefeld. As the name Helmholtz may already suggest, the physical sciences held a prominent place there. In particular, Dr Lippert, my physics teacher, gave me a splendid grounding in the fundamentals of physics. Although he was suffering from cancer, he instructed us with unbelievable care and attention. He seemed, in particular, to have a very soft spot for me. On Saturday afternoons, I would go to his house, where he immediately brought me into the family circle; afterwards we went through the details of the current lessons. You can well understand the deep sadness we students felt at the death of our physics teacher. I have to thank him for my special liking for physics. On 2 March 1932 I passed the Matriculation examination; the commendation states "Gottfried Möllenstedt wants to be an engineer".

\* Translated by T. Mulvey.

## II. GAINING TECHNICAL KNOW-HOW AS A STUDENT APPRENTICE

At first I wanted to study aircraft construction. That meant that practical apprenticeship of several months was necessary. I worked in the Anker Works and in the Adler Works in Bielefeld from April to December in 1932, in the section locksmith's shop, the planing shop, the milling and grinding shops, the carpentry shop, hardening shop, the smithy and iron foundry. In the summer semester of 1933 I began the study of marine and aircraft technology in Danzig. Applying myself to constructional methods, materials science, machine drawing and hull-shape drawing gave me much pleasure as well as constituting a sound preparation for the technology of experimental physics. However, I soon came to the conclusion that it was the field of experimental physics that would give the best chance of carrying out actual experiments. So, on the advice of a member of the staff who was an *Assistent*, i.e. one who assisted the Professor, I transferred in May 1934 to the Faculty of Mathematics and Physics in the Kossel Institute.

## III. A BROAD EDUCATION IN PHYSICS WITH PROFESSOR WALTER KOSSEL

Here I learned what experimental physics is all about and how one carries out research. I had the good fortune soon to be appointed as an *Assistent* myself and to experience the full sweep of physics. To be sure, the Institute was not large, but work was going on in the most varied of research fields. I was fascinated by the X-ray interference patterns from lattice sources. These Kossel lines were to become well known throughout the world. The Borrmann anomalous transparency for X-rays in single crystals arose out of these X-ray investigations. Single crystal pulling and the growth of solid bodies, i.e. solid state physics, formed the principal research area of the Institut. With total love and devotion, Kossel (Fig. 1) carried out electron diffraction experiments at voltages of 30–800 kV with, at that time, home-made high-voltage generators. The physics of thin films and electron microscopy was held in high esteem and vigorously promoted by Kossel and his research student, Georg Hass. The best education, however, for me was my role as technician in the great experimental lectures given by Kossel. These held sway over all other events in the Institute at that time. I emphasize these aspects of my multi-faceted learning phase as being particularly important for my ability to do independent research. If one wants to invent and develop something new, one must have observed and experienced how a productive and inspired researcher draws out new concepts. Walter Kossel is to be numbered among the great physicists of his time.



**FIG. 1** Walter Kossel, 1888–1956.

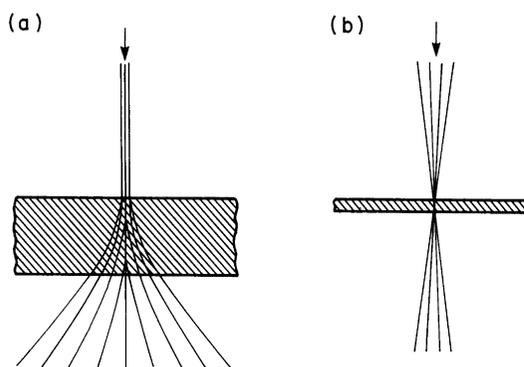
#### IV. INFLUENCE OF THE WORK OF KIKUCHI AND CONVERGENT BEAM DIFFRACTION

As a preparatory phase to my later discovery of electron biprism interference, I must include my diploma work in convergent beam electron diffraction. The wonderful electron diffraction patterns of Seichi Kikuchi are, of course, now well known. A parallel monochromatic electron beam that penetrates a single crystal several hundred nanometres thick or is reflected at the surface of a single crystal produces on a photographic plate bright and dark lines characteristic of the single crystal and frequently bounded by bright and dark bands. The explanation of these phenomena occupied a central position in the theoretical activity of the Danzig Institute. It would be no exaggeration to say that no name was uttered so frequently in the Danzig Institute as that of the Japanese physicist Kikuchi (Fig. 2). Now it is well known that the Kikuchi patterns are a statistical result of the interaction of electrons with the single crystal. One day, Kossel commissioned me to take up this field as



**FIG. 2** Seichi Kikuchi and Gottfried Möllenstedt in Tokyo in 1961.

a diploma investigation, in order to clarify the fundamental physical processes involved. I used an extremely thin single crystal, which I could irradiate with a convergent electron beam formed by electron optical means (Fig. 3). The technical details of how my diploma investigation was carried out cannot be discussed here. However, I must emphasize that I learnt as much from this experiment as I did from the theory! As a researcher, I learnt how Kossel



**FIG. 3** Electron diffraction. (a) Kikuchi method. (b) Convergent beam method of Kossel and Möllenstedt.

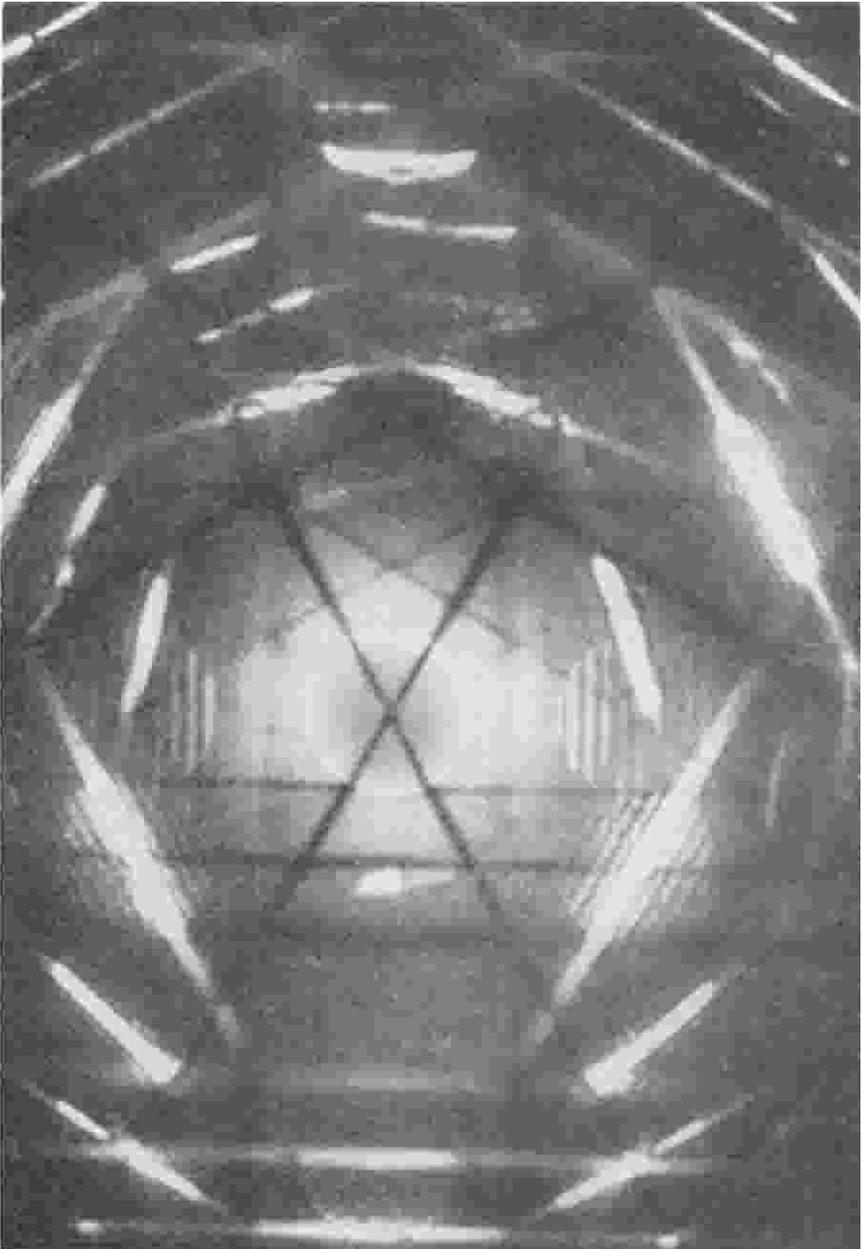
went about things. Peer intently, and learn the existing state of knowledge. And then ask oneself "How can we go on further to new results?" These were to be provided later by the convergent beam method (Fig. 4).

But now, to come to the discovery of electron biprism interference. Already in the Kossel Institute there had been much discussion about the interaction of waves with matter. The concept of "coherence length"; for example, was already playing a role in the years around 1938. The book "Theory and Practice of Electron Diffraction" by the English physicists G. P. Thomson and W. Cochrane had been discussed in minute detail. From light optics I knew all about interferometry from the Danzig experiments. However, no one dared think about interferometry with electron waves at this time.

At about the time (1952) that I took up the Chair of Applied Physics at the University of Tübingen, there came the news of Marton's idea of arranging several crystals in series so as to split and then reunite a monochromatic electron beam, as shown in Fig. 5. The overlapping wave trains are coherent. The experiments were carried out by Arol Simpson at the National Bureau of Standards. They were fiendishly tedious and, as it turned out later, lacked reproducibility. We realized in Tübingen, on the basis of my experience with electron beams and single crystals, that this kind of interferometry could never provide a solution for electron beam interferometry.

## V. ELECTRON-OPTICAL EXPERIMENTS WITH BRÜCHE, SCHERZER AND MAHL

Now I come to the moment of inspiration for the idea of using an electron biprism for electron beam interferometry. From 1949 until 1952, I worked in the Süddeutsche Laboratorien in Mosbach-Baden where I had been appointed by Professor Brüche as a Section Leader after I had given up working as an independent manufacturer of simplified electron microscopes. At Mosbach, I worked with electrostatic microscopes of the type AEG/Zeiss/EM7 shown in Fig. 6. I was able to produce dark-field images in the Zeiss EM7 by fixing a fine tungsten wire over the hole in the objective aperture mounted in the back focal plane of the objective lens. In this way, one could record, in a single micrograph, as shown in Fig. 7, both the bright-field and the dark-field image. After a prolonged period of operation, however, this novel aperture arrangement gave rise to imperfections in the final image. As a consequence of the electron bombardment of the aperture, a now well-known "contamination" layer was formed on its surface. This led to electrical charging-up effects at the wire. The most remarkable effect was that one frequently got "double" images, as may be seen in Fig. 8. Figure 9 shows a light micrograph, taken in a reflection optical microscope, of the  $5\ \mu\text{m}$



**FIG. 4** Convergent beam electron diffraction pattern of Muscovite taken at 50 kV. Target diameter 120 nm.