

Fundamentals of High-Resolution Transmission Electron Microscopy

S. Horiuchi



1994

North-Holland

Amsterdam - London - New York - Tokyo

Contents

Preface	
Chapter 1. Introduction	1
1. Background for yielding an electron microscope	1
1.1. Discovery of electrons	1
1.2. Material wave and electron diffraction	1
2. Invention and progress of a transmission electron microscope	2
2.1. Trial production of an electron lens	2
2.2. Mechanical improvement of TEMs	4
2.3. Development of specimen preparation techniques	4
2.4. Progress of analytical functions	5
3. Definition of resolving power	7
3.1. Resolving power of an optical microscope	7
3.2. Resolving power of a TEM with diffraction contrast	9
3.3. Resolving power of a TEM with phase contrast	10
Chapter 2. Electron lens	11
1. Geometry and function of an electron lens	11
1.1. Practical arrangement of electron lenses in a TEM	11
1.1.1. Imaging mode	11
1.1.2. Diffraction mode	11
1.2. Magnetic lens	12
1.2.1. Locus of electrons in a magnetic lens	12
1.2.2. Approximation in a bell-shaped magnetic field	14
1.2.3. Functional parameters of a practical electron lens	15
2. Lens aberration	16
2.1. Spherical aberration	16
2.1.1. Origin of the spherical aberration	16
2.1.2. Focal length of an optical convex lens	18
2.1.3. Spherical aberration constant	19
2.1.4. Blurring of images due to spherical aberration	19
2.1.5. Dependence of the spherical aberration on the strength of the lens excitation	20
2.2. Chromatic aberration	21
2.2.1. Dependence on the stability of the accelerating voltage	21
2.2.2. Dependence on the stability of the lens current	22
2.3. Astigmatism	23
2.3.1. Asymmetry of the magnetic field	23

2.3.2. Second-order astigmatism	23
2.3.3. Mechanical correction of astigmatism	25
2.3.4. Caustic plane	25
2.3.5. Third-order astigmatism	26
2.4. Minimization of lens aberrations	27
2.4.1. Analysis of the magnetic field by a finite element method	27
2.4.2. Design of an electron lens	28
References	29
 Chapter 3. Generation and control of an electron beam	31
1. Electron gun	31
1.1. Thermal emission gun	31
1.1.1. Definition of the brightness	31
1.1.2. Hair-pin-type W filament	31
1.1.3. Brightness with a thermal emission gun	33
1.1.4. LaB ₆ cathode	33
1.2. Field emission gun	35
1.2.1. Schottky effect	35
1.2.2. Pure field emission	35
1.2.3. Temperature and field emission	37
2. Control of the electron beam	38
2.1. Necessity of beam control	38
2.1.1. Parallel beam	38
2.1.2. Electron probe	38
2.2. Condenser lens	39
2.2.1. Beam diameter and illumination angle	39
2.2.2. Double condenser lens	40
2.2.3. Triple condenser lens	40
2.2.4. Critical illumination and Köhler illumination	42
References	42
 Chapter 4. Properties of an electron wave	43
1. Description of a wave	43
1.1. Wave equation	43
1.1.1. Derivation of a wave equation	43
1.1.2. Plane wave and spherical wave	44
1.2. Periodic wave	45
1.2.1. Sinusoidal wave	45
1.2.2. Expression by a complex notation	46
1.2.3. Intensity of a wave	47
1.3. Wave equation for an electron wave	48
1.3.1. Schrödinger equation	48
1.3.2. Physical meaning of a wave function	48

1.3.3. Propagation of electron waves in free space	48
2. Interference of electron waves	49
2.1. Synthesis of waves	49
2.1.1. Synthesis of waves by geometrical drawing	49
2.1.2. Wave packet	51
2.2. Conditions for the occurrence of interference	54
2.2.1. Length and period of interference	54
2.2.2. Parallelism and monochromatism of electron waves	55
2.3. Degree of coherence	56
2.3.1. Correlation between two optical beams	56
2.3.2. Coherence, partial coherence and incoherence	57
2.3.3. Intensity of interference fringes	59
2.3.4. Measurement of interference fringes	60
3. Fraunhofer diffraction	61
3.1. Amplitude of diffracted waves	62
3.1.1. Formation of a wave front by secondary waves	62
3.1.2. Huygens–Fresnel principle	62
3.1.3. Kirchhoff diffraction integral	64
3.1.4. Application of the diffraction integral	65
3.2. Diffraction by a single hole	66
3.2.1. Diffraction by a rectangular hole	66
3.2.2. Diffraction by a circular hole	68
3.2.3. Babinet principle	70
3.3. Diffraction by a number of holes	71
3.3.1. Irregularly arrayed holes	71
3.3.2. Regularly arrayed holes – grating	72
4. Fresnel diffraction	74
4.1. Formation of Fresnel diffraction patterns	74
4.1.1. Characteristics of Fresnel fringes	74
4.1.2. Underfocus and overfocus	75
4.2. Intensity of Fresnel fringes	76
4.2.1. Opaque specimen	76
4.2.2. Cornu spiral	77
4.2.3. Partly transmittable specimen	79
4.2.4. Effect of the cross-sectional shape of the specimen	81
4.2.5. Effect of illumination angle	82
References	83
Chapter 5. Electron diffraction	85
1. Kinematical diffraction	85
1.1. Scattering of electron waves by atoms	85
1.1.1. Scattering of electron waves by a potential field	85
1.1.2. Born approximation	87
1.1.3. Atomic scattering factor	87

1.2.	Diffraction of electron waves by a crystal	91
1.2.1.	Crystal structure factor	91
1.2.2.	Reciprocal lattice, Laue condition and Bragg condition	91
1.2.3.	Ewald sphere	93
1.2.4.	Diffraction intensity	94
1.2.5.	Extinction rule	96
1.3.	Relativistic effect	97
1.3.1.	Wavelength correction	97
1.3.2.	Relationship with the accelerating voltage	98
1.4.	Elastic and inelastic scattering	99
1.4.1.	Thermal diffuse scattering	99
1.4.2.	Appearance of Kikuchi lines	100
1.4.3.	Effect on HRTEM images	102
2.	Dynamical diffraction	102
2.1.	Necessity of the theory of dynamical diffraction	103
2.1.1.	Scattering from a column	103
2.1.2.	Extinction distance	104
2.2.	Column approximation method (Howie–Whelan method)	105
2.2.1.	Two-beam approximation	105
2.2.2.	Diffraction intensity	106
2.3.	Eigenvalue method (Bethe method)	107
2.3.1.	Fundamental equation	107
2.3.2.	Two-beam approximation	108
2.3.3.	Diffraction intensity	109
2.3.4.	Three-beam approximation	111
2.3.5.	Many-beam effect	113
2.4.	Multi-slice method (Cowley–Moody method)	114
2.4.1.	Phase object	115
2.4.2.	Propagation function	116
2.4.3.	Fundamental equation	117
2.4.4.	Effect of the deviation from a zone-axis orientation	120
2.4.5.	Some examples of calculated diffraction intensities and phases for thin crystals	122
	References	126
	Chapter 6. Imaging mechanism for transmission electron microscopy	127
1.	Diffraction contrast images	127
1.1.	Formation mechanism of image contrast	129
1.1.1.	Bright- and dark-field images	129
1.1.2.	Equal thickness fringe and bend contour	130
1.1.3.	Anomalous transmission	132
1.2.	Observation of lattice defects	134
1.2.1.	Diffraction amplitude	134

1.2.2. Image of a dislocation	135
1.2.3. Selection of diffraction conditions	137
2. Crystal lattice images	138
2.1. Formation of a lattice image	139
2.1.1. Phase contrast	139
2.1.2. Wave-optical imaging mechanism	140
2.1.3. Lattice fringes by two-beam interference	141
2.1.4. Many-beam lattice image	142
2.2. Wave aberration	145
2.2.1. Effect of spherical aberration	145
2.2.2. Effect of defocus	145
2.2.3. Aberration function	145
2.2.4. Dependence on spatial frequency	146
3. Crystal structure images	148
3.1. Image contrast of a weak-phase object	148
3.1.1. Weak-phase object approximation	148
3.1.2. Phase contrast transfer function	149
3.1.3. Scherzer condition	150
3.1.4. Resolution limit due to spherical aberration	150
3.1.5. Similarity to a phase-difference microscope	151
3.2. Structure images from crystalline specimens	151
3.2.1. Extension of weak-phase object approximation	151
3.2.2. Transfer function modified by the effect of dynamical diffraction	152
3.2.3. Reversal of image contrast	153
3.2.4. Information obtained from structure images	154
3.2.5. Electro-optical conditions to obtain a structure image	158
3.2.6. Possible research targets of HRTEM observation	160
3.2.6.1. Structure analysis	160
3.2.6.2. Identification of lattice imperfections	165
4. Observation of very weakly scattering substances	166
4.1. Phase contrast image of an amorphous thin film	167
4.1.1. Thon curve	167
4.1.2. Granular structure	168
4.2. Image contrast of single atoms	168
4.2.1. Bright-field imaging method	168
4.2.2. Dark-field imaging method	170
References	171
Chapter 7. Resolution limit due to the coherence degree of electron waves	175
1. Optical functions for evaluating the degree of coherence	175
1.1. Mutual intensity and coherence degree	175

1.1.1. van Cittert–Zernike theorem	175
1.1.2. Intensity of an image by two-beam interference	177
1.1.3. Hopkins formula	178
1.2. Response function of an optical system	179
1.2.1. Propagation of the mutual intensity	179
1.2.2. Response function and pupil function	179
1.3. Transmission cross coefficient	181
1.3.1. Introduction of the transmission cross coefficient	181
1.3.2. Role of the transmission cross coefficient	182
1.3.3. Symmetry in the image intensity	183
2. Information transfer and resolution limit for HRTEM	184
2.1. Optical functions for transmission electron microscopy	185
2.1.1. Incorporation of the effect of chromatic aberration	185
2.1.2. Effective electron source	186
2.1.3. Transmission cross coefficient for high-resolution observation	186
2.2. Modulation function for the image intensity	188
2.2.1. Effect of the chromatic aberration	188
2.2.2. Effect of the convergence of the incident beam	189
2.3. Modulation of the phase contrast transfer function	190
2.3.1. Modulation function for the weak-phase object	190
2.3.2. Resolution limit due to chromatic aberration	192
2.3.3. Resolution limit due to beam convergence	193
References	193
 Chapter 8. How to observe and analyze HRTEM images	195
1. Observation of many-beam lattice images	195
1.1. Preparation techniques of thin specimens	195
1.1.1. Classification	195
1.1.2. Crushing method	195
1.1.3. Ion-milling method	197
1.1.4. Electro-polishing method	199
1.2. Operation of TEMs	199
1.2.1. Procedure for observing a many-beam lattice image	199
1.2.2. Setting of a zone-axis orientation	201
1.2.3. Beam alignment	203
1.2.4. Specimen contamination and how to get high-quality vacuum	205
1.3. Specimen damage due to electron irradiation	206
1.3.1. Phenomena of specimen damage	206
1.3.2. Ionization damage	207
1.3.3. Knock-on damage	208

1.3.4. Critical dose and resolution limit for irradiation-sensitive specimens	209
1.3.5. How to reduce the damage rate	210
2. Image analysis	211
2.1. Computer simulation of an HRTEM image	211
2.1.1. Computer program for calculating an HRTEM image	211
2.1.2. Examination at each stage of the computer calculation	214
2.1.3. Calculation of images for lattice defects	215
2.2. Optical diffraction	217
2.2.1. Optical diffractometer	217
2.2.2. Array of optical diffraction spots	217
2.2.3. Optical diffractograms from some crystalline specimens	218
2.2.3.1. Identification of substances	219
2.2.3.2. Analysis of a zone axis	220
2.2.3.3. Determination of the extinction rule	220
2.2.3.4. Extraction of a regularity in a lattice image	221
2.2.4. Optical diffractograms from an amorphous thin film	222
2.2.5. Correction of astigmatism by means of an optical diffractogram	223
2.3. Resolving power	224
2.3.1. Point resolving power	225
2.3.2. Information limit	225
2.3.3. Line resolving power	225
2.4. Measurement of optical parameters defining the resolving power	226
2.4.1. Defocus amount	226
2.4.2. Spherical aberration constant	227
2.4.3. Fluctuation of focus	227
2.4.4. Convergent angle	229
2.5. Image processing	231
2.5.1. Restoration of HRTEM images	231
2.5.2. Extraction of information	232
3. Analysis of electron diffraction patterns	233
3.1. Bragg diffraction spots	233
3.1.1. Indexing of diffraction spots	233
3.1.1.1. Ring patterns	233
3.1.1.2. Spot patterns	234
3.1.2. Identification of substances	235
3.1.3. Analysis of crystal orientation	235
3.1.4. Determination of extinction rule and lattice parameters	237
3.2. Kikuchi lines	239
3.2.1. Accurate measurement of crystal orientation	239

3.2.2. Measurement of accelerating voltage	240
References	240
Chapter 9. Applications of HRTEM for analyzing high- T_c superconductors	243
1. Y-based superconductors	243
1.1. Fundamental structure	243
1.2. Structure of a twin boundary in $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$	245
1.2.1. Introduction	245
1.2.2. Experimental	246
1.2.3. Results and discussion	246
1.3. Stacking faults in Ag-sheathed $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ tapes	250
1.3.1. Introduction	250
1.3.2. Experimental	251
1.3.3. Superconductive properties	252
1.3.4. Microstructures	253
1.3.5. Discussion	257
1.4. Distribution of oxygen atoms in a $\text{YBa}_2\text{Cu}_3\text{O}_{6.4}$	258
1.4.1. Introduction	258
1.4.2. Experimental	258
1.4.3. Results and discussion	259
1.5. Visualization of oxygen atoms in $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ by UHR-HVEM	265
1.5.1. Introduction	265
1.5.2. Computer simulation of images	265
1.5.3. Observation of real images	268
1.5.4. Discussion on image contrast	269
2. Bi-based superconductors	273
2.1. Fundamental structure	273
2.1.1. Introduction	273
2.2. Modulated structure of the 2212 phase	275
2.2.1. Introduction	275
2.2.2. Superstructure analyzed by computer simulation of HRTEM images	276
2.2.3. Simple description of the modulated structure	279
2.3. Phase transitions from the 2201 to the 2223 via the 2212 phase	280
2.3.1. Introduction	280
2.3.2. Experimental	281
2.3.3. Results and discussion	281
2.4. Microstructure evolution in the 2223 phase	291
2.4.1. Introduction	291
2.4.2. Experimental	292

2.4.3. Results	292
2.4.4. Discussion	299
References	301
 Appendix	305
1. Commonly used physical constants and conversion factors	305
2. Relationships among the accelerating voltage E , the wavelength λ and the velocity v_e of electrons	306
3. Geometrical relations for crystals	306
3.1. Relationships between the interplanar spacing and the lattice parameters	306
3.2. Angles between two lattice planes	307
4. The extinction rule of electron diffraction spots and the space group of crystals	308
5. Atomic scattering factors for electrons, f	316
5.1. Atomic scattering amplitudes for electrons, obtained by the self-consistent field calculation	316
5.2. Atomic scattering amplitudes for electrons, calculated based on a Thomas-Fermi-Dirac statistical model	318
6. Computer simulation of HRTEM	321
6.1. Source list for calculating HRTEM image intensity (CIHRTEM)	321
6.2. Data list for CIHRTEM	334
 Index	335