

V.M. Agranovich V.L. Ginzburg

Crystal Optics with Spatial Dispersion, and Excitons

Second Corrected and Updated Edition

With 46 Figures

Springer-Verlag
Berlin Heidelberg New York Tokyo 1984

PHYSIKALISCHE BIBLIOTHEK
FACHBEREICH 5
TECHNISCHE HOCHSCHULE
DARMSTADT

T/3140

Contents

1. Introduction	1
1.1 Basic Problems of Crystal Optics	1
1.2 Excitons and Polaritons	10
2. The Complex Dielectric-Constant Tensor $\varepsilon_{ij}(\omega, k)$ and Normal Waves in a Medium	18
2.1 The Tensor $\varepsilon_{ij}(\omega, k)$ and Its Properties	18
2.1.1 The Electromagnetic Field Equations and the Introduction of the Tensor $\varepsilon_{ij}(\omega, k)$	18
2.1.2 General Properties of the Tensor $\varepsilon_{ij}(\omega, k)$	27
2.1.3 The Approximation of Classical Crystal Optics. The Tensor $\varepsilon_{ij}(\omega, k)$ in an Isotropic Medium	37
2.2 Normal Electromagnetic Waves in a Medium	42
2.2.1 Wave Equation and Dispersion Equation	42
2.2.2 Transverse and Longitudinal Waves, "Fictitious" Longitudinal Waves and "Polarization Waves". Real, Coulomb and Mechanical Excitons	47
a) Coulomb Excitons: Longitudinal and "Fictitious" Longitudinal Waves	51
b) Coulomb Excitons: "Polarization Waves"	54
2.2.3 Multiple Roots of the Dispersion Equation	57
2.2.4 Separating the Transverse Field E_{\perp} and the Tensor $\varepsilon_{\perp, ij}$	62
2.2.5 The Dispersion Relations for a Complex Refractive Index. Inequalities for the Region of Transparency	66
2.3 Energy Relations and Other Equations for Waves in an Anisotropic Medium	72
2.3.1 The Law of Energy Conservation in the Electrodynamics of Media Displaying Spatial Dispersion	72
2.3.2 Quadratic Functions of the Normal Wave Amplitudes	81
a) The Conservation of Energy for a Field	83
b) The Vector of Group Velocity	84
c) The Conservation of Momentum for a Field	93
d) Some Applications of the Poynting Theorem	94
e) The Boundary Conditions for a Gyrotropic Medium	95

f) The Boundary Conditions for the Optically Nonlinear Medium with the Center of Inversion	97
g) Spatial Dispersion and Orthogonality of Normal Waves	99
h) The Reciprocity Theorem	100
2.3.3 Certain Theorems for Ray and Wave Propagation in a Medium	103
3. The Tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ in Crystals	108
3.1 The Tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ for Crystals	108
3.1.1 Introduction of the Tensor $\varepsilon_{ij}(\omega, \mathbf{k})$	108
3.1.2 Weak Spatial Dispersion	117
3.2 The Tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ for Crystals of Various Classes	122
3.2.1 Gyrotropic Crystals	122
3.2.2 Nongyrotropic Crystals	129
4. Spatial Dispersion in Crystal Optics	136
4.1 Gyrotropic Crystals	136
4.1.1 Normal Waves in Gyrotropic Crystals	137
4.1.2 A New Wave Near the Absorption Lines in a Gyrotropic Medium	145
4.2 Nongyrotropic Crystals	149
4.2.1 Normal Waves	149
4.2.2 Isotropic Medium and New Waves Near Dipole Absorption Lines	150
a) Group-Velocity Vector	165
4.3 Cubic Nongyrotropic Crystals	167
4.3.1 Optical Anisotropy: Dipole Transitions	167
4.3.2 Optical Anisotropy: Quadrupole Transitions	171
4.3.3 Classification of the States of 'Mechanical' Excitons with $k = 0$ and Selection Rules for Quadrupole Transitions ...	174
4.3.4 New Waves Near Quadrupole Absorption Lines. Longitudinal Waves	185
4.4 Influence of Mechanical Stresses and External Electric and Magnetic Fields	188
4.4.1 Anisotropy of the Optical Properties and Selection Rules in the Presence of External Influences	188
4.4.2 The Explicit Dependence of the Tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ on the Strength of Weak External Fields	192
a) The Magnetic-Field Inversion Effect	196
4.4.3 Influence of Magnetic and Electric Fields on Cadmium Sulfide (CdS) Crystals	198
a) Quadratic Magneto-Stark Effect	202
4.5 Boundary Conditions in the Case of Spatial Dispersion Near a Separate Resonance (Absorption Line)	205

4.5.1	The Tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ Near an Isolated Resonance	208
4.5.2	Examples	211
a)	Dielectric Tensor for Isotropic Nongyrotropic Media	216
4.5.3	Boundary Conditions	218
a)	The Transition ("Dead") Layer Problem	224
b)	ABC for Molecular Crystals and the Surface Current Appearance	228
4.5.4	Reflected and Refracted Waves Near Dipole and Quadrupole Transition Frequencies in a Nongyrotropic Crystal	233
a)	The Law of Energy Conservation	236
4.5.5	Reflected and Refracted Waves Near a Dipole Transition Frequency in a Gyrotropic Crystal	238
4.5.6	The Influence of a Nonhomogeneous Subsurface Layer on Light Reflection Near Exciton-Absorption Bands	242
4.5.7	Transmission of Light Through a Plane-Parallel Plate (Nongyrotropic Medium)	244
4.5.8	Transmission of Light Through a Plane-Parallel Plate (Gyrotropic Medium)	248
a)	The Integral Absorption	247
4.6	Experimental Investigations of Spatial Dispersion in Crystals	253
4.6.1	Gyrotropic Crystals	253
4.6.2	Nongyrotropic Crystals	257
5.	Surface Excitons and Polaritons	271
5.1	Polaritons at the Interface of Isotropic Media	271
5.1.1	Dispersion of Surface Waves for Lossless Media	274
5.1.2	General Case	274
5.1.3	Surface Polaritons for Layered Structures	275
a)	The Brewster Waves	278
b)	Propagation of Surface Waves with Damping	279
c)	Reflection, Diffraction and Refraction of Surface Waves at Surface Boundaries	280
5.2	Spectra of Surface Polaritons in Anisotropic Crystals	283
5.3	Transition-Layer Effects in Surface Polariton Spectra	289
5.3.1	Transition Layers with High Electric Conductivity	290
5.3.2	Transition Layers in the Presence of a Resonance with the Surface Polariton	294
a)	The Effective Boundary Conditions	294
b)	Polariton Spectrum Splitting for TH Surface Waves	295
c)	TE Surface Waves in the Transition Layers' Resonance Region	298
5.3.3	Effect of Surface Roughness (Irregularities) on the Path Length of Surface Polaritons	301

a) Surface Polariton Scattering in the Vicinity of Phase-Transition Points	302
5.4 Effect of Spatial Dispersion on the Spectra of Surface Polaritons, and Additional ("New") Surface Waves	303
5.4.1 Surface Electromagnetic Waves at the Right-Left Gyrotropic Crystal Interface	311
5.5 Experimental Investigations of Surface Polaritons	314
5.5.1 The Attenuated Total Reflection (ATR) Method	315
5.5.2 The Ruled Diffraction-Grating Method	319
5.5.3 Raman Scattering of Light by Surface Polaritons	319
5.5.4 Surface Polariton Propagation Over Long Distances. Crystal Optics of Surfaces	323
5.5.5 The Inelastic Low-Energy Electron Scattering (ILES) Method	324
5.5.6 Nonlinear Surface Electromagnetic Waves	325
6. Microscopic Theory. Calculation of the Tensor $\varepsilon_{ij}(\omega, \mathbf{k})$	328
6.1 General Expressions for $\varepsilon_{ij}(\omega, \mathbf{k})$	328
6.1.1 Quantum-Mechanical Derivation of $\varepsilon_{ij}(\omega, \mathbf{k})$	328
6.1.2 Contribution of the Exciton States to the Tensor $\varepsilon_{ij}(\omega, \mathbf{k})$	337
a) Refractive Index Near Exciton Resonances of Anisotropic Crystals	340
6.2 Mechanical Excitons and the Tensor $\varepsilon_{ij}(\omega, \mathbf{k})$ in Molecular Crystals and in the Classical Oscillator Model	342
6.2.1 Molecular Crystals. Mechanical Excitons	342
6.2.2 Calculating the Dielectric-Constant Tensor of Molecular Crystals by the Local-Field Method	348
a) Davydov Splitting	351
6.2.3 The Oscillator Model	353
6.3 Absorption	356
6.3.1 The Absorption Mechanism. Absorption in a First Approximation	356
6.3.2 Absorption of Normal Electromagnetic Waves in the Vicinity of an Exciton Transition Frequency	361
6.3.3 The Long-Wavelength Edge of the Exciton Absorption Bands: Raman and Brillouin-Mandelstam Scattering of Polaritons	363
6.4 Raman Scattering of Light and X Rays Accompanied by Exciton Production. Influence of Spatial Dispersion on Energy Losses, and on Cherenkov and Transition Radiation of Charged Particles	367
6.4.1 Raman Scattering of X Rays Accompanied by Exciton Production	368
6.4.2 Raman Scattering of Light by Polaritons	373
a) Raman Scattering of Light by Bulk Polaritons	373

b) General Expression for the Scattering Intensity	378
c) The Scattering Cross Section by Polariton. Linewidth .	380
d) Raman Scattering of Light by Surface Polaritons	383
e) Compensation Effect	387
f) Broken Symmetry Method	389
g) Coherent Anti-Stokes Raman Spectroscopy (CARS) ..	393
6.4.3 Energy Losses and Cherenkov Radiation of a Charge Travelling with Uniform Motion Through a Medium Displaying Spatial Dispersion. Transition Radiation	396
7. Conclusion	402
Appendix	406
A.1 Crystal-Symmetry Notation	406
A.2 Information from Space Group Theory	409
A.2.1 Classification of the States of Mechanical Excitons	414
Notation	417
References	419
Subject Index	437