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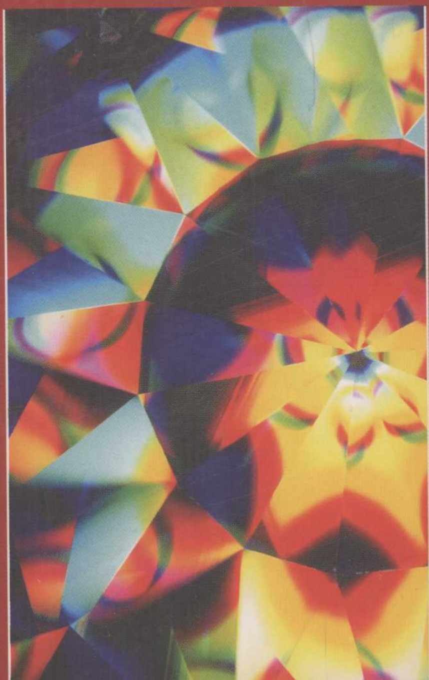
Optical Properties of Solids

固体的光学性质



M. Fox

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Preface

This book is about the way light interacts with solids. The beautiful colours of gemstones have been valued in all societies, and metals have been used for making mirrors for thousands of years. However, the scientific explanations for these phenomena have only been given in relatively recent times. Nowadays, we build on this understanding and make use of rubies and sapphires in high power solid state lasers. Meanwhile, the arrival of inorganic and organic semiconductors has created the modern optoelectronics industry. The onward march of science and technology therefore keeps this perennial subject alive and active.

The book is designed for final year undergraduates and first year graduate students in physics. At the same time, I hope that some of the topics will be of interest to students and researchers of other disciplines such as engineering or materials science. It evolved from a final year undergraduate course in condensed matter physics given as part of the Master of Physics degree at Oxford University. In preparing the course I became aware that the discussion of optical phenomena in most of the general solid state texts was relatively brief. My aim in writing was therefore to supplement the standard texts and to introduce new subjects that have come to the fore in the last 10–20 years.

Practically all textbooks on this subject are built around a number of core topics such as interband transitions, excitons, free electron reflectivity, and phonon polaritons. This book is no exception. These core topics form the backbone for our understanding of the optical physics, and pave the way for the introduction of more modern topics. Much of this core material is well covered in the standard texts, but it can still benefit from the inclusion of more recent experimental data. This is made possible through the ever-improving purity of optical materials and the now widespread use of laser spectroscopy.

The overall plan of the subject material is summarized in Fig. 1. The flow diagram shows that some of the chapters can be read more or less independently of the others, on the assumption that the introductory material in Chapters 1 and 2 has been fully assimilated. I say 'more or less' here because it does not really make sense, for example, to try to understand nonlinear optics without a firm grasp of linear optics. The rest of the chapters have been arranged into groups, with their order following a certain logical progression. For example, knowledge of interband absorption is required to understand quantum wells, and is also needed to explain certain details in the reflectivity spectra of metals. Similarly, molecular materials provide an intuitive introduction to the concept of configuration diagrams, which are required for the understanding of colour centres and luminescent impurities.

The inclusion of recent developments in the subject has been one of the main priorities motivating this work. The chapters on semiconductor quantum

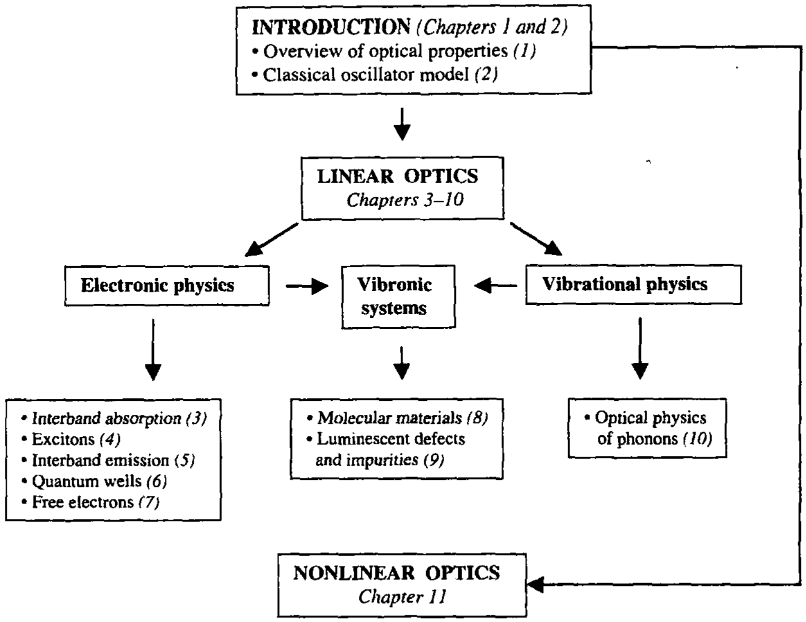

Fig. 1 Scheme of the subject topics covered in this book. The numbers in brackets refer to the chapters where the subject material is developed.

wells, molecular materials, and nonlinear optics will not be found in most of the standard texts. Other new topics such as the Bose–Einstein condensation of excitons are included alongside traditional subject material. Furthermore, it is my deliberate intention to illustrate the physics with up-to-date examples of optical technology. This provides an interesting modern motivation for traditional topics such as colour centres and also helps to emphasize the importance of the solid state devices.

Throughout the book I have understood the term 'optical' in a wider sense than its strict meaning referring to the visible spectral region. This has allowed me to include discussions of infrared phenomena such as those due to phonons and free carriers, and also the properties of insulators and metals in the ultraviolet. I have likewise taken the scope of the word 'solid' beyond the traditional emphasis on crystalline materials such as metals, semiconductors and insulators. This has allowed me to include both 'soft condensed matter' materials, such as polymers, and also glasses, which are not solids in the strict sense.

The process of relating measured optical phenomena to the electronic and vibrational properties of the material under study can proceed in two ways. We can work forwards from known electronic or vibrational physics to predict the results of optical experiments, or we can work backwards from experimental data to the microscopic properties. An example of the first approach is to use the free electron theory to explain why metals reflect light, while an example of the second is to use absorption or emission data to deduce the electron level structure of a crystal. Textbooks such as this one inevitably tend to work forwards from the microscopic properties to the measured data, even though an experimental scientist would probably be working in the other direction.

The book presupposes that the reader has a working knowledge of solid state physics at the level appropriate to a third year undergraduate, such as that found in H.M. Rosenberg's *The Solid State* (Oxford University Press, third edition, 1988). This puts the treatment at about the same as, or at a slightly higher level, than that given in the *Introduction to Solid State Physics* by Charles Kittel. The book also necessarily presupposes a reasonable knowledge of electromagnetism and quantum theory. Classical and quantum arguments are used interchangeably throughout, and the reader will need to revise their own favourite texts on these subjects if any of the material is unfamiliar. Three appendices are included to provide a succinct summary of the principal results from band theory, electromagnetism and quantum theory that are presupposed.

The text has been written in a tutorial style, with worked examples in most chapters. A collection of exercises is provided at the end of each chapter, with solutions at the end of the book. The exercises follow the presentation of the material in the chapter, and the more challenging ones are identified with an asterisk.

Acknowledgements

I would like to acknowledge the many people who have helped in various ways in the production of this book. Pride of place goes to Soenke Adlung at Oxford University Press, for instigating and editing the Oxford Masters Series, and to my series co-authors, Stephen Blundell and John Singleton, who have offered many useful ideas on how to approach the project. I would also like to thank Anja Tschoertner and Richard Lawrence at the Press for handling the day-to-day administration of the manuscript and producing the book.

Numerous colleagues have helped to clarify my understanding of certain specialist topics and have made comments on the text. Among these I would like to single out Dr Simon Martin and Dr Paul Lane for their critical reading of the chapter on molecular materials. I am, of course, grateful to my teachers, who enthused me with the subject, and to the students who have taken the course and tried out some of the problems.

I would like to thank Dr Robert Taylor for providing me with unpublished data for use in Figs 5.3, 6.13 and 6.17, Adam Ashmore for taking the data in Figs 5.6 and 5.13, and Drs Gero von Plessen and Andrew Tomlinson for the data presented in Fig. 4.5. I would also like to thank Excerpta Medica Inc. for granting permission to reproduce Figs 1.6, 4.4, 4.7, 7.13, 8.10, 8.16, 8.19, 9.6, and 10.11; the Optical Society of America for Fig. 1.7; the American Physical Society for Figs 3.4, 3.7, 4.3, 7.2, 7.3, 7.7, 7.11, 10.7, and 10.14; Springer-Verlag for Figs 3.9 and 3.12; the Physical Society of Japan for Fig. 4.9; the American Institute of Physics for Figs 5.8, 6.10, 8.9, 8.17, and 11.9; Taylor and Francis Ltd for Figs 6.9 and 6.12; the Institute of Physics, Polish Academy of Sciences, for Fig. 6.16; Verlag der Zeitschrift für Naturforschung for Fig. 8.13; Kluwer Academic/Plenum Publishers for Figs 9.5, 9.15, and 11.7; Academic Press for Figs 9.9 and 10.5; Penwell Corp. for Fig. 9.11; and the IEEE for Fig. 11.10.

Finally, I would like to thank the Royal Society for supporting me as a University Research Fellow during the years in which most of the book has been written.

M.F.

Sheffield
January 2001

目　录

1　导论 ······ 1
1.1　光学过程的分类 ······ 1
1.2　光学系数 ······ 2
1.3　复折射率和介电常数 ······ 5
1.4　光学材料 ······ 8
1.4.1　晶体、绝缘体和半导体 ······ 8
1.4.2　玻璃 ······ 11
1.4.3　金属 ······ 12
1.4.4　分子材料 ······ 13
1.4.5　掺杂玻璃和绝缘体 ······ 14
1.5　固态的光物理特性 ······ 15
1.5.1　晶体对称性 ······ 16
1.5.2　电子能带 ······ 18
1.5.3　振动带 ······ 19
1.5.4　态密度 ······ 19
1.5.5　非定域态和集体激发 ······ 19
1.6　微观模型 ······ 20
2　经典传播 ······ 25
2.1　光密介质中光的传播 ······ 25
2.1.1　原子振荡 ······ 26
2.1.2　振动振荡 ······ 28
2.1.3　自由电子振荡 ······ 29
2.2　偶极振荡模型 ······ 29
2.2.1　Lorentz 振荡 ······ 29
2.2.2　多重共振 ······ 34
2.2.3　与实验结果的对照 ······ 37
2.2.4　局域场修正 ······ 39
2.2.5　Kramers-Kronig 关系 ······ 40
2.3　色散 ······ 40
2.4　光学各向异性:双折射 ······ 42
3　带间吸收 ······ 49
3.1　带间跃迁 ······ 49
3.2　直接吸收跃迁率 ······ 51

3.3 直接带隙半导体的带间吸收 …… 54
3.3.1 带间吸收的原子物理学 …… 54
3.3.2 直接带隙 III-V 半导体的能带结构 …… 56
3.3.3 联合态密度 …… 57
3.3.4 带边吸收的频率特性 …… 58
3.3.5 Franz-Keldysh 效应 …… 60
3.3.6 磁场中的带间吸收 …… 61
3.4 直接带隙半导体的带边吸收 …… 63
3.5 带边上的带间吸收 …… 66
3.6 吸收谱的测量 …… 68
3.7 半导体光电探测器 …… 69
3.7.1 光电二极管 …… 69
3.7.2 光电导器件 …… 71
3.7.3 光伏器件 …… 71
4 激子 …… 76
4.1 激子的概念 …… 76
4.2 自由激子 …… 77
4.2.1 结合能和半径 …… 77
4.2.2 激子吸收 …… 79
4.2.3 GaAs 中自由激子的实验数据 …… 80
4.3 外场中的自由激子 …… 82
4.3.1 电场 …… 82
4.3.2 磁场 …… 83
4.4 高密度自由激子 …… 84
4.5 Frenkel 激子 …… 86
4.5.1 稀有气体晶体 …… 87
4.5.2 碱金属卤化物 …… 87
4.5.3 分子晶体 …… 88
5 发光学 …… 92
5.1 固体的发光 …… 92
5.2 带间发光 …… 95
5.2.1 直接带隙材料 …… 95
5.2.2 非直接带隙材料 …… 96
5.3 光致发光 …… 98
5.3.1 激发和弛豫 …… 98
5.3.2 低载流子密度 …… 100
5.3.3 简并 …… 101
5.3.4 光致发光的光谱 …… 102

5.4 电致发光 …… 103
5.4.1 电致发光器件的原理 …… 103
5.4.2 发光二极管 …… 106
5.4.3 二极管激光器 …… 107
6 半导体量子阱 …… 115
6.1 量子约束结构 …… 115
6.2 量子阱的生长和结构 …… 117
6.3 电子能级 …… 119
6.3.1 变量分离 …… 119
6.3.2 有限深势阱 …… 119
6.3.3 无限深势阱 …… 121
6.4 光学吸收和激子 …… 124
6.4.1 选择定则 …… 125
6.4.2 二维吸收 …… 127
6.4.3 实验数据 …… 129
6.4.4 量子阱中的激子 …… 130
6.5 量子约束 Stark 效应 …… 131
6.6 光发射 …… 133
6.7 子带间跃迁 …… 135
6.8 Bloch 振荡 …… 135
6.9 量子点 …… 136
6.9.1 半导体掺人玻璃 …… 137
6.9.2 自组织 III-V 量子点 …… 138
7 自由电子 …… 143
7.1 等离子体反射率 …… 143
7.2 自由载流子导电性 …… 145
7.3 金属 …… 147
7.3.1 Drude 模型 …… 148
7.3.2 金属中的带间跃迁 …… 151
7.4 掺杂半导体 …… 154
7.4.1 自由载流子的反射率和吸收 …… 154
7.4.2 非本征吸收 …… 158
7.5 等离体子 …… 160
8 分子材料 …… 165
8.1 分子材料导论 …… 166
8.2 共轭分子的电子态 …… 167
8.3 分子光谱 …… 169
8.3.1 电子-振动跃迁 …… 170

8.3.2 分子构型图 …… 171
8.3.3 Frank-Condon 原理 …… 173
8.3.4 实验光谱 …… 176
8.4 芳香烃 …… 177
8.5 共轭高分子 …… 179
8.6 有机光电材料 …… 181
9 发光中心 …… 186
9.1 振动吸收和发射 …… 186
9.2 色心 …… 189
9.3 离子晶体中的顺磁杂质 …… 192
9.3.1 晶体场效应和振动耦合 …… 192
9.3.2 稀土离子 …… 193
9.3.3 过渡金属离子 …… 195
9.4 固态激光器和光学放大器 …… 196
9.5 磷光体 …… 199
10 光子 …… 204
10.1 红外活性光子 …… 204
10.2 极化固体的红外反射率和吸收 …… 206
10.2.1 经典振子模型 …… 206
10.2.2 Lyddane-Sachs-Teller 关系 …… 209
10.2.3 Restrahlen …… 210
10.2.4 点阵吸收 …… 212
10.3 电磁耦子 …… 214
10.4 极化子 …… 215
10.5 非弹性光散射 …… 218
10.5.1 非弹性光散射的原理 …… 218
10.5.2 Raman 散射 …… 220
10.5.3 Brillouin 散射 …… 222
10.6 光子寿命 …… 222
11 非线性光学 …… 227
11.1 非线性极化率张量 …… 227
11.2 光学非线性的物理起源 …… 230
11.2.1 非共振非线性 …… 230
11.2.2 共振非线性 …… 234
11.3 二阶非线性 …… 236
11.3.1 非线性混频 …… 237
11.3.2 晶体对称性 …… 239
11.3.3 相位匹配 …… 241

11.4 三阶非线性效应 …… 243
11.4.1 三阶现象回顾 …… 243
11.4.2 各向同性三阶非线性介质 …… 247
11.4.3 半导体中的共振非线性 …… 249
附录A 电介质的电磁理论 …… 255
A.1 电磁场和 Maxwell 方程 …… 255
A.2 电磁波 …… 258
附录B 辐射吸收和发射 …… 263
B.1 爱因斯坦系数 …… 263
B.2 量子跃迁率 …… 266
B.3 选择定则 …… 269
附录C 能带理论 …… 271
C.1 金属、半导体和绝缘体 …… 271
C.2 近自由电子模型 …… 273
C.3 能带结构实例 …… 276
附录D 半导体 p-i-n 二极管 …… 280
习题解答 …… 282
参考文献 …… 290
符号 …… 295
索引 …… 297

Contents

1 Introduction 1

1.1 Classification of optical processes 1
1.2 Optical coefficients 2
1.3 The complex refractive index and dielectric constant 5
1.4 Optical materials 8
1.4.1 Crystalline insulators and semiconductors 8
1.4.2 Glasses 11
1.4.3 Metals 12
1.4.4 Molecular materials 13
1.4.5 Doped glasses and insulators 14
1.5 Characteristic optical physics in the solid state 15
1.5.1 Crystal symmetry 16
1.5.2 Electronic bands 18
1.5.3 Vibronic bands 19
1.5.4 The density of states 19
1.5.5 Delocalized states and collective excitations 19
1.6 Microscopic models 20

2 Classical propagation 25

2.1 Propagation of light in a dense optical medium 25
2.1.1 Atomic oscillators 26
2.1.2 Vibrational oscillators 28
2.1.3 Free electron oscillators 29
2.2 The dipole oscillator model 29
2.2.1 The Lorentz oscillator 29
2.2.2 Multiple resonances 34
2.2.3 Comparison with experimental data 37
2.2.4 Local field corrections 39
2.2.5 The Kramers–Kronig relationships 40
2.3 Dispersion 40
2.4 Optical anisotropy: birefringence 42

3 Interband absorption 49

3.1 Interband transitions 49
3.2 The transition rate for direct absorption 51
3.3 Band edge absorption in direct gap semiconductors 54
3.3.1 The atomic physics of the interband transitions 54
3.3.2 The band structure of a direct gap III–V semiconductor 56
3.3.3 The joint density of states 57

3.3.4 The frequency dependence of the band edge absorption 58
3.3.5 The Franz–Keldysh effect 60
3.3.6 Band edge absorption in a magnetic field 61
3.4 Band edge absorption in indirect gap semiconductors 63
3.5 Interband absorption above the band edge 66
3.6 Measurement of absorption spectra 68
3.7 Semiconductor photodetectors 69
3.7.1 Photodiodes 69
3.7.2 Photoconductive devices 71
3.7.3 Photovoltaic devices 71

4 Excitons **76**
4.1 The concept of excitons 76
4.2 Free excitons 77
4.2.1 Binding energy and radius 77
4.2.2 Exciton absorption 79
4.2.3 Experimental data for free excitons in GaAs 80
4.3 Free excitons in external fields 82
4.3.1 Electric fields 82
4.3.2 Magnetic fields 83
4.4 Free excitons at high densities 84
4.5 Frenkel excitons 86
4.5.1 Rare gas crystals 87
4.5.2 Alkali halides 87
4.5.3 Molecular crystals 88

5 Luminescence **92**
5.1 Light emission in solids 92
5.2 Interband luminescence 95
5.2.1 Direct gap materials 95
5.2.2 Indirect gap materials 96
5.3 Photoluminescence 98
5.3.1 Excitation and relaxation 98
5.3.2 Low carrier densities 100
5.3.3 Degeneracy 101
5.3.4 Photoluminescence spectroscopy 102
5.4 Electroluminescence 103
5.4.1 General principles of electroluminescent devices 103
5.4.2 Light emitting diodes 106
5.4.3 Diode lasers 107

6 Semiconductor quantum wells **115**
6.1 Quantum confined structures 115
6.2 Growth and structure of semiconductor quantum wells 117
6.3 Electronic levels 119
6.3.1 Separation of the variables 119
6.3.2 Infinite potential wells 119
6.3.3 Finite potential wells 121
6.4 Optical absorption and excitons 124

6.4.1 Selection rules 125
6.4.2 Two-dimensional absorption 127
6.4.3 Experimental data 129
6.4.4 Excitons in quantum wells 130
6.5 The quantum confined Stark effect 131
6.6 Optical emission 133
6.7 Intersubband transitions 135
6.8 Bloch oscillators 135
6.9 Quantum dots 136
6.9.1 Semiconductor doped glasses 137
6.9.2 Self-organized III–V quantum dots 138

7 Free electrons **143**
7.1 Plasma reflectivity 143
7.2 Free carrier conductivity 145
7.3 Metals 147
7.3.1 The Drude model 148
7.3.2 Interband transitions in metals 151
7.4 Doped semiconductors 154
7.4.1 Free carrier reflectivity and absorption 154
7.4.2 Impurity absorption 158
7.5 Plasmons 160

8 Molecular materials **165**
8.1 Introduction to molecular materials 166
8.2 Electronic states in conjugated molecules 167
8.3 Optical spectra of molecules 169
8.3.1 Electronic–vibrational transitions 170
8.3.2 Molecular configuration diagrams 171
8.3.3 The Franck–Condon principle 173
8.3.4 Experimental spectra 176
8.4 Aromatic hydrocarbons 177
8.5 Conjugated polymers 179
8.6 Organic optoelectronics 181

9 Luminescence centres **186**
9.1 Vibronic absorption and emission 186
9.2 Colour centres 189
9.3 Paramagnetic impurities in ionic crystals 192
9.3.1 The crystal field effect and vibronic coupling 192
9.3.2 Rare earth ions 193
9.3.3 Transition metal ions 195
9.4 Solid state lasers and optical amplifiers 196
9.5 Phosphors 199

10 Phonons **204**
10.1 Infrared active phonons 204
10.2 Infrared reflectivity and absorption in polar solids 206
10.2.1 The classical oscillator model 206
10.2.2 The Lyddane–Sachs–Teller relationship 209

10.2.3 Restrahlen 210
10.2.4 Lattice absorption 212
10.3 Polaritons 214
10.4 Polarons 215
10.5 Inelastic light scattering 218
10.5.1 General principles of inelastic light scattering 218
10.5.2 Raman scattering 220
10.5.3 Brillouin scattering 222
10.6 Phonon lifetimes 222

11 Nonlinear optics **227**
11.1 The nonlinear susceptibility tensor 227
11.2 The physical origin of optical nonlinearities 230
11.2.1 Non-resonant nonlinearities 230
11.2.2 Resonant nonlinearities 234
11.3 Second-order nonlinearities 236
11.3.1 Nonlinear frequency mixing 237
11.3.2 Crystal symmetry 239
11.3.3 Phase matching 241
11.4 Third-order nonlinear effects 243
11.4.1 Overview of third-order phenomena 243
11.4.2 Isotropic third-order nonlinear media 247
11.4.3 Resonant nonlinearities in semiconductors 249

A Electromagnetism in dielectrics **255**
A.1 Electromagnetic fields and Maxwell's equations 255
A.2 Electromagnetic waves 258

B Quantum theory of radiative absorption and emission **263**
B.1 Einstein coefficients 263
B.2 Quantum transition rates 266
B.3 Selection rules 269

C Band theory **271**
C.1 Metals, semiconductors and insulators 271
C.2 The nearly free electron model 273
C.3 Example band structures 276

D Semiconductor p-i-n diodes **280**

Solutions to exercises **282**

Bibliography **290**

Symbols **295**

Index **297**

1

Introduction

Light interacts with matter in many different ways. Metals are shiny, but water is transparent. Stained glass and gemstones transmit some colours, but absorb others. Other materials such as milk appear white because they scatter the incoming light in all directions.

In the chapters that follow, we will be looking at a whole host of these optical phenomena in a wide range of solid state materials. Before we can begin to do this, we must first describe the way in which the phenomena are classified, and the coefficients that are used to quantify them. We must then introduce the materials that we will be studying, and clarify in general terms how the solid state is different from the gas and liquid phase. This is the subject of the present chapter.

1.1 Classification of optical processes 1
1.2 Optical coefficients 2
1.3 The complex refractive index and dielectric constant 5
1.4 Optical materials 8
1.5 Characteristic optical physics in the solid state 15
1.6 Microscopic models 20

1.1 Classification of optical processes

The wide-ranging optical properties observed in solid state materials can be classified into a small number of general phenomena. The simplest group, namely **reflection**, **propagation** and **transmission**, is illustrated in Fig. 1.1. This shows a light beam incident on an optical medium. Some of the light is reflected from the front surface, while the rest enters the medium and propagates through it. If any of this light reaches the back surface, it can be reflected again, or it can be transmitted through to the other side. The amount of light transmitted is therefore related to the reflectivity at the front and back surfaces and also to the way the light propagates through the medium.

The phenomena that can occur while light propagates through an optical medium are illustrated schematically in Fig. 1.2.

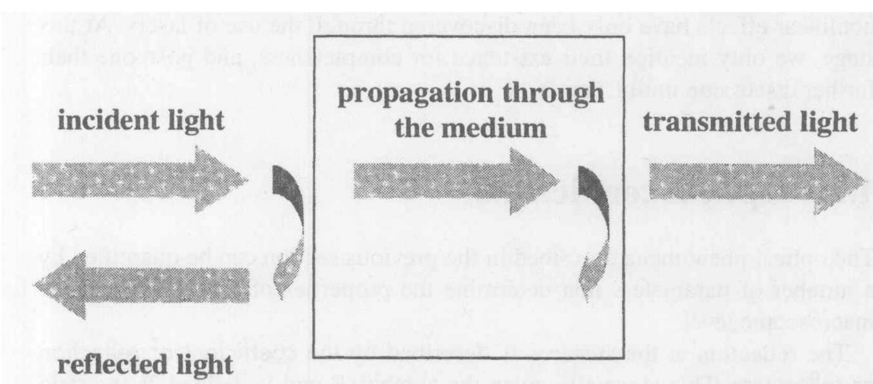

Fig. 1.1 Reflection, propagation and transmission of a light beam incident on an optical medium.

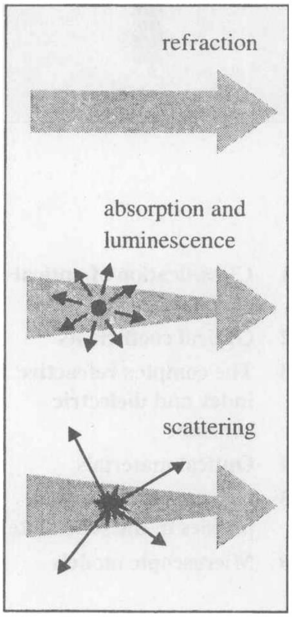

Fig. 1.2 Phenomena that can occur as a light beam propagates through an optical medium. Refraction causes a reduction in the velocity of the wave, while absorption causes attenuation. Luminescence can accompany absorption if the excited atoms re-emit by spontaneous emission. Scattering causes a redirection of the light. The diminishing width of the arrow for the processes of absorption and scattering represents the attenuation of the beam.

Refraction causes the light waves to propagate with a smaller velocity than in free space. This reduction of the velocity leads to the bending of light rays at interfaces described by Snell's law of refraction. Refraction, in itself, does not affect the intensity of the light wave as it propagates.

Absorption occurs during the propagation if the frequency of the light is resonant with the transition frequencies of the atoms in the medium. In this case, the beam will be attenuated as it progresses. The transmission of the medium is clearly related to the absorption, because only unabsorbed light will be transmitted. Selective absorption is responsible for the colouration of many optical materials. Rubies, for example, are red because they absorb blue and green light, but not red.

Luminescence is the general name given to the process of spontaneous emission of light by excited atoms in a solid state material. One of the ways in which the atoms can be promoted into excited states prior to spontaneous emission is by the absorption of light. Luminescence can thus accompany the propagation of light in an absorbing medium. The light is emitted in all directions, and has a different frequency to the incoming beam.

Luminescence does not always have to accompany absorption. It takes a characteristic amount of time for the excited atoms to re-emit by spontaneous emission. This means that it might be possible for the excited atoms to dissipate the excitation energy as heat before the radiative re-emission process occurs. The efficiency of the luminescence process is therefore closely tied up with the dynamics of the de-excitation mechanisms in the atoms.

Scattering is the phenomenon in which the light changes direction and possibly also its frequency after interacting with the medium. The total number of photons is unchanged, but the number going in the forward direction decreases because light is being re-directed in other directions. Scattering therefore has the same attenuating effect as absorption. The scattering is said to be **elastic** if the frequency of the scattered light is unchanged, or **inelastic** if the frequency changes in the process. The difference in the photon energy in an inelastic scattering process has to be taken from the medium if the frequency increases or given to the medium if the frequency decreases.

A number of other phenomena can occur as the light propagates through the medium if the intensity of the beam is very high. These are described by **nonlinear optics**. An example is frequency doubling, in which the frequency of part of a beam is doubled by interaction with the optical medium. These nonlinear effects have only been discovered through the use of lasers. At this stage, we only mention their existence for completeness, and postpone their further discussion until Chapter 11.

1.2 Optical coefficients

The optical phenomena described in the previous section can be quantified by a number of parameters that determine the properties of the medium at the macroscopic level.

The reflection at the surfaces is described by the **coefficient of reflection** or **reflectivity**. This is usually given the symbol R and is defined as the ratio of the reflected power to the power incident on the surface. The **coefficient**