Photons and Nonlinear Optics
Photons and Nonlinear Optics

Revised and Enlarged Edition

David N. Klyshko

Professor of Quantum Radiophysics
Department of Physics, Moscow State University,
Moscow, USSR

Translated from the Russian by
Yuri Sviridov
Contents

Preface xv
List of Notation xxi

CHAPTER 1 INTRODUCTION 1

1.1 Parametric Scattering 1
   The Spectrum of Parametric Scattering 2
   Coherent Scattering 4
   Zero Fluctuations of the Vacuum 5
   The Effective Temperature of Parametric Scattering 6
   The Number of Photons per Mode and the Brightness of Light 9
   The Brightness of Parametric Scattering 13
   Parametric Super luminescence 14
   Tuning Curves 17
   Integral Intensities of Parametric Scattering 19

1.2 Scattering by Polaritons 21
   Polaritons and the Dispersion Law 21
   Comparison of Parametric Scattering and Scattering by Polaritons 23
   The Intensity of Scattering by Polaritons 24
   The Frequency-Angular Spectrum of Scattering by Polaritons 25
   Normal and Anomalous Dispersion 26
   The Linearization Effect 28

1.3 Four-Photon Scattering and Nonlinear Spectroscopy 28
   Hyperparametric Scattering 29
   Cascade Hyperparametric 31
   Hyper-Raman Scattering 31
   Biphoto Radiation in Raman Scattering 32
   Resonance Fluorescence 33
## CONTENTS

Two-Photon Resonance Luminescence 33  
Nonlinear Spectroscopy 34  

### 1.4 The Basic Concepts of Statistical Optics 34  
- The Analytic Signal 35  
- Random Intensity 36  
- Correlation Functions 39  
- Temporal Coherence 40  
- Spatial Coherence 43  
- Coherence Volume and the Degeneracy Factor 44  
- Photocount Statistics, Mandel's Formula 45  
- Photo Bunching 50  
- The Correlation of Intensities 51  

### 1.5 Multiphoton Effects: A Historic and Bibliographic Outline 53  
- The Stone (Pre-Laser) Age 54  
- The Laser Age 56  
- Three-Photon Parametric Scattering 57  
- Other Types of Scattering 59  
- Hyperparametric Scattering 60  
- Quantum Optics 62  

## CHAPTER 2 SOME INFORMATION ON QUANTUM MECHANICS AND STATISTICAL PHYSICS 65  

- Passing from a Classical Description to a Quantum Description 65  
- The Quantization of the equations of Motion 65  
- Choice of representation 68  
- Comparison of Theory and Experiment 68  
- The Experimental Procedure 70  

### 2.2 The Dirac Notation and the Geometric Interpretation of Quantum Mechanics 71  
- Functions as Vectors 71  
- The Transformation of Functions and Tensors 73  
- Unity Decomposition 75  
- Various Representations 76
## CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unitary Transformations</td>
<td>78</td>
</tr>
<tr>
<td>The Connection with Physics</td>
<td>79</td>
</tr>
<tr>
<td>Time as a Parameter</td>
<td>80</td>
</tr>
<tr>
<td>Quantum Correlation Functions</td>
<td>83</td>
</tr>
<tr>
<td>Mixed States and the Density Matrix</td>
<td>83</td>
</tr>
<tr>
<td><strong>2.3 The Interaction Representation and Perturbation Theory</strong></td>
<td>85</td>
</tr>
<tr>
<td>The Interaction Representation</td>
<td>86</td>
</tr>
<tr>
<td>Other Representations</td>
<td>88</td>
</tr>
<tr>
<td>Perturbation Theory for the Scattering Operator</td>
<td>89</td>
</tr>
<tr>
<td>Transition Probability</td>
<td>91</td>
</tr>
<tr>
<td>The Rate of Transition</td>
<td>93</td>
</tr>
<tr>
<td><strong>2.4 Fluctuation-Dissipation Theorems</strong></td>
<td>94</td>
</tr>
<tr>
<td>The Linear Fluctuation-Dissipation Theorem</td>
<td>95</td>
</tr>
<tr>
<td>The Spectral Form</td>
<td>99</td>
</tr>
<tr>
<td>The Symmetry of Moments and Susceptibilities</td>
<td>100</td>
</tr>
<tr>
<td>The Quadratic FDT</td>
<td>101</td>
</tr>
<tr>
<td>The Approximate Cubic FDT</td>
<td>104</td>
</tr>
<tr>
<td><strong>2.5 Relaxation and Kinetic Equations</strong></td>
<td>106</td>
</tr>
<tr>
<td>The Kinetic Equation for the Density matrix</td>
<td>107</td>
</tr>
<tr>
<td>The Kinetic Equations for Mean Values</td>
<td>108</td>
</tr>
<tr>
<td>The Kinetic Equations for Populations</td>
<td>111</td>
</tr>
<tr>
<td>Spontaneous and Stimulated Transitions</td>
<td>112</td>
</tr>
</tbody>
</table>

## CHAPTER 3 THE PRINCIPLES OF QUANTUM OPTICS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>3.1 Canonical Variables of the Electromagnetic Field</strong></td>
<td>115</td>
</tr>
<tr>
<td>Dynamical Equations of the Field</td>
<td>115</td>
</tr>
<tr>
<td>The Spatial Fourier Transform of the Field</td>
<td>117</td>
</tr>
<tr>
<td>Canonical Variables of the Field</td>
<td>119</td>
</tr>
<tr>
<td>Canonical Equations of the Field and Green Function</td>
<td>120</td>
</tr>
<tr>
<td>The Role of the Dimensions of the Quantization Box</td>
<td>121</td>
</tr>
<tr>
<td><strong>3.2 Quantization of the Electromagnetic Field</strong></td>
<td>122</td>
</tr>
<tr>
<td>The Field Hamiltonian</td>
<td>122</td>
</tr>
<tr>
<td>Commutators of Field Operators</td>
<td>124</td>
</tr>
</tbody>
</table>
The Free Field 127
Decomposition of Field Operators into Fourier’s Frequency Integral 128

3.3 Possible States of the Field 130
Energy and Coherent States 130
The Multimode Field 133
The Properties of Basis States 135
Mixed States 137
The Characteristic Function 139
The Dynamics of Field States 141
The Scattering Matrix of the Field 145
Coordinate, Momentum, and Squeezed States 145

3.4 Photon and Photoelectric Statistics 152
Photon Statistics 153
Photon Bunching and Antibunching 158
Photoelectron Statistics 163

3.5 The Atom-Quantized Field Interaction 168
Absorption and Radiation Probabilities 168
Spontaneous Radiation 170
The Interaction of Stationary Systems 172
The Spectral Representation 176
Equilibrium Systems. The Fluctuation-Dissipation Theorem 178

3.6 Green’s Function and the Quantization of the Macroscopic Field in a Medium 180
Maxwell’s Phenomenological Equations 180
Green’s Function for a Field in a Medium 182
Normal Waves 184
The Dispersion Law 184
The Passage to the kt Representation 185
Field Quantization in a Medium 187
The wr Representation 190

CHAPTER 4 THERMAL RADIATION IN THE LINEAR APPROXIMATION 193
4.1 The Statistics of an Equilibrium Field in Free Space 194
   The Equilibrium Statistical Operator 194
   The Characteristic Function 195
   Photon Statistics 195

4.2 Fluctuations of a Macroscopic Field in Matter 198
   The Two-Stage Calculation Procedure 198
   The Direct Path 199
   The \(kt\) Representation 201
   The Observation Problem 201
   Fluctuations of the Field in an Isotropic Medium 202

4.3 The Thermal Radiation of Heated Bodies 205
   Application of the Fluctuation-Dissipation Theorem 206
   Kirchhoff's Law 208

4.4 The Generalized Kirchhoff Law 210
   The Input and Output Field 210
   The Scattering Matrix 211
   Vector Notation 213
   Nyquist's Derivation of the Generalized Kirchhoff Law 214
   Special Cases 216
   The Higher Moments of Thermal Radiation 217
   The Thermal Field in Perturbation Theory 219
   Langevin's Derivation of the Generalized Kirchhoff Law 220

4.5 The Kinetic Equation Derivation of the Generalized Kirchhoff's Law 221
   Thermal Radiation and the Kinetic Equation 221
   The Kinetic Equation for the \(\chi\) Function 222
   Equations for Moments 227
   Quantum Amplifier Noises 228

4.6 Thermal Radiation in the Far Field 229
   The Far Field and the \(a_k\) Operators 229
   The Correlation Function 231
   Measuring the Scattering Matrix Modulus 232
   The Microscopic Model 233
CONTENTS

The Coherence Volume 235
Photon Counting 237
The Phenomenological Model 238

4.7 The Interference of Intensities 240
The Probability of Photocount Coincidence 241
The Role of the Detector Volume 243
Interpretation of the Effect 246

CHAPTER 5 THE INFLUENCE OF THE ANHARMONISM OF MATTER ON THERMAL RADIATION 249

5.1 Multiphoton Transitions and the Higher Field Moments 250
Anharmonism and the Correlation of Different-Frequency Modes 250
Equilibrium and Nonequilibrium Problems 251
The Two-Photon Paradox 252

5.2 Calculation of Field Moments by Means of Perturbation Theory 253
The Relationship Between Output and Input Moments 255
The General Properties of Transformation 257
The Higher Corrections 259

5.3 The Third Moment of the Thermal Field 260
The Connection with the Two-Time Correlation Function of Interpretation of the Effect 263
Estimating the Cube of the Field 265
The Detection Problem 267

5.4 Kirchhoff’s Two-Photon Law 270
The Effective Kinetic Equation 270
The First Moment 272
The Second Moments 273
The Fourth Moments 276
Kirchhoff’s Two-Photon Law 277
The Comparison with One-Photon Thermal Radiation 278
<table>
<thead>
<tr>
<th>Section</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.1</td>
<td>The Rate of Biphoton Generation</td>
<td>286</td>
</tr>
<tr>
<td></td>
<td>The Third Order of Perturbation Theory</td>
<td>286</td>
</tr>
<tr>
<td></td>
<td>The Effective Hamiltonian</td>
<td>289</td>
</tr>
<tr>
<td></td>
<td>The Probability of Biphoton Creation</td>
<td>291</td>
</tr>
<tr>
<td></td>
<td>The Differential Rate of Biphoton Creation</td>
<td>292</td>
</tr>
<tr>
<td></td>
<td>The Law of Conservation of Momentum</td>
<td>294</td>
</tr>
<tr>
<td>6.2</td>
<td>The Integral Intensity of Parametric Scattering</td>
<td>295</td>
</tr>
<tr>
<td></td>
<td>The Low Frequency-Resolution Detector</td>
<td>296</td>
</tr>
<tr>
<td></td>
<td>Light Brightness in Parametric Scattering</td>
<td>298</td>
</tr>
<tr>
<td></td>
<td>The Low Angular-Resolution Detector</td>
<td>299</td>
</tr>
<tr>
<td></td>
<td>Allowing for the Nonparallelism of the Group and Phase Velocities</td>
<td>301</td>
</tr>
<tr>
<td>6.3</td>
<td>The Form of the Spectral Line of Parametric Scattering</td>
<td>302</td>
</tr>
<tr>
<td></td>
<td>Coherence Length</td>
<td>303</td>
</tr>
<tr>
<td></td>
<td>k Spectroscopy</td>
<td>305</td>
</tr>
<tr>
<td></td>
<td>The Effective Linewidth</td>
<td>307</td>
</tr>
<tr>
<td></td>
<td>The Degeneracy Case</td>
<td>307</td>
</tr>
<tr>
<td></td>
<td>The Influence of the Pump Spectrum on the Signal Spectrum</td>
<td>308</td>
</tr>
<tr>
<td></td>
<td>The Gaussian Pump Beam</td>
<td>310</td>
</tr>
<tr>
<td>6.4</td>
<td>Field Statistics and the Metrological Applications of Parametric Scattering</td>
<td>313</td>
</tr>
<tr>
<td></td>
<td>The Heisenberg Equations for Field Operators</td>
<td>315</td>
</tr>
<tr>
<td></td>
<td>Spontaneous Scattering</td>
<td>317</td>
</tr>
<tr>
<td></td>
<td>A Standard Photon Generator</td>
<td>318</td>
</tr>
<tr>
<td></td>
<td>The Rate of Coincidences for a Gaussian Pump</td>
<td>321</td>
</tr>
<tr>
<td></td>
<td>The Absolute Radiometer</td>
<td>325</td>
</tr>
<tr>
<td></td>
<td>Measurement Errors</td>
<td>327</td>
</tr>
<tr>
<td>6.5</td>
<td>Parametric Superluminescence</td>
<td>328</td>
</tr>
<tr>
<td></td>
<td>Monochromatic Pumping</td>
<td>330</td>
</tr>
<tr>
<td></td>
<td>The Characteristic Function</td>
<td>331</td>
</tr>
<tr>
<td></td>
<td>The One-Dimensional Model: Wqz and Kz Representations</td>
<td>334</td>
</tr>
</tbody>
</table>
CONTENTS

Modulated Pumping and the Slowly Varying Amplitude Representation 338

6.6 Scattering by Polariton 342
Description of Polariton Scattering with a Linear Fluctuation–Dissipation Theorem 343
Application of the Nonlinear Fluctuation-Dissipation Theorem 346
The Unipolar Approximation 351
Kirchhoff’s Law for Parametric and Polariton Scattering 353

CHAPTER 7 HYPERPARAMETRIC AND RAMAN SCATTERING 359

7.1 Nonresonance Hyperparametric Scattering 359
The Centrosymmetric Medium 360
Hyperparametric Scattering in Piezocrystals 361
The Intensity of Hyperparametric Scattering in Piezocrystals 366
The Statistics of Cascade Hyperparametric Scattering 367

7.2 Resonance Hyperparametric Scattering and Raman Scattering 368
The Equation for the Slowly Varying Field Operators 369
Vacuum Noise 371
Molecular Noise 372

7.3 The Correlation of Stokes and Anti-Stokes Radiation in Raman Scattering 373
The Classical Model 373
The Phenomenological Description 375
Correlation in the One-Dimensional Approximation 378
The Coincidence Rate 381
The Gaussian Pump 384
Estimation of the Number of Coincidences 388

APPENDIX THE GREEN FUNCTION AND TYPES OF FIELD POLARIZATION IN AN ANISOTROPIC ABSORBING MEDIUM 391
CONTENTS

Diagonalization and Inversion of Non-Hermitian Matrices 391
Eigenvectors and Eigenvalues of the Green Tensor 395
The Eigenvectors for Tensors $\pi \varepsilon^{-1}$ and $\varepsilon^{-1} \pi$ 396
The Anisotropic Angles 399

References 401

Subject Index 411
In Memory of R.V. Khokhlov
This book was originally conceived as an introduction to quantum optics for college students with an initial grounding in quantum mechanics and for experimental physicists, most of whom have successfully forgotten it. That is why Chapter 2 sets out, in contracted form, some of the basic concepts of quantum mechanics and statistics. To avoid putting off the reader by an avalanche of formulas, Chapter 1 contains a simplified description of parametric light scattering, one of the most spectacular phenomena of quantum optics. This phenomenon is readily visible to the naked eye as an iridescent ring; it is attributed to the spontaneous decay of photons into photon pairs with fractional frequencies due to the optical nonlinearity of the transparent material involved. The introductory chapter gives a description of similar effects and provides some background information on their discovery, along with a summary of the basic concepts of quantum optics.

The transition from classical electrodynamics to quantum electrodynamics is discussed in Chapter 3, which also describes a classification of possible statistical states of the field, the statistics of photons in these states, and the associated photoelectron statistics.

The following four chapters and the Appendix, some fifty percent of the book, are based on the results of the author's own research. Chapters 4 and 5 are devoted to the one- and two-photon thermal radiation of matter in equilibrium, respectively. In these chapters, general relationships are established between spontaneous and stimulated effects that generalize Kirchhoff's law for nongeometric and nonlinear optics and include higher field moments. Analogous relationships are also used in the next two chapters, which are concerned with inelastic scattering processes. Chapter 6 contains a more detailed treatment of parametric and polariton scattering than Chapter 1. Chapter 7
presents a phenomenological description of four-photon (hyperparametric) scattering and the associated phenomenon of spontaneous coherent anti-Stokes scattering. Finally, the Appendix contains a definition of the spectral Green function and describes various types of polarization for the field in an anisotropic absorbing medium.

The feature of this book that distinguishes it from the well-known fundamental monographs on quantum and statistical optics by Glau- ber, Klauder and Sudarshan, Louisell, Loudon, Lax, Scully, Haken, Perina, and Akhmanov and Chirkin, is its emphasis on multimode fields with correlating different-frequency modes, notably on their phenomenological description and practical methods of generating them.

The title of this book may seem contradictory at first sight; after all, the term “nonlinear optics” is normally associated with powerful laser radiation involving vast quantities of photons, so it would seem that there is no need to take the photon structure of light into account. Indeed, the vast majority of effects in nonlinear (and linear, for that matter) optics are adequately described by the semiclassical theory of radiation, in which the electromagnetic field obeys Maxwell’s classical equations and only the behavior of matter is quantum in character.

However, certain optical phenomena are difficult, if not impossible, to interpret through the use of only classical concepts; a consistent theory should describe both atoms and light proceeding from the principles of quantum mechanics. Besides, clear photon representations are convenient for the qualitative description and classification of many optical effects. For instance, the effect of light frequency doubling or addition during the passage of light through a transparent crystal can be seen as the result of many elementary processes, in each of which two photons of incident light merge into a single photon with combined energy and frequency. The reverse process—the decay of the photon incident on the crystal into a pair of photons with lower energies—is apparently also possible. Such processes account for the phenomenon of the parametric scattering of light. In Raman scattering, the incident photon turns into a lower-frequency photon called the Stokes photon and a quantum of the excitation of matter (e.g., in the case of vibrational excitation, a phonon). In addition, an incident photon may combine with a thermal phonon to form a higher-frequency anti-Stokes photon. In two-photon absorption, two incident photons may excite the atom, the molecule, or the crystal.
Conversely, an excited atom may pass into the ground state after emitting a pair of photons.

Of the great variety of nonlinear optical effects discovered over the past twenty years, we shall, for the most part, examine only those phenomena which are related to the emission of photons in pairs (for brevity, we shall refer to photon pairs that arise simultaneously as "biphotons"). The emission of photons in pairs (as well as in threes, fours, etc.) is closely associated with the optical nonlinearity of the material involved; this emission graphically demonstrates the quantum properties of light. We shall also discuss the photon bunching effect (the Brown–Twiss effect), which is not affected by the nonlinearity of the material and does not require field quantization for an explanation.† This kind of bunching is almost random; photon pairs are encountered only twice as frequently in ordinary light as in the chaotic Poisson flux of dust particles (this is due to the wave nature of light). It is worth noting that two-photon absorption, in contrast, leads to a uniform distribution of photons in light passing through matter (the antibunching effect) and, in lasers, the nonlinearity of the working substance (the saturation effect) distributes photons chaotically, in a Poisson distribution.

Biphotons should be radiated not only in the phenomena of parametric scattering and two-photon decay, which were observed in the laser era of the history of optics, but also in the long-familiar spontaneous Raman scattering. As we will show elsewhere in this book, anti-Stokes photons will only be emitted paired with Stokes photons at low temperatures of the scattering material. Four-photon, or hyperparametric, scattering, which differs from three-photon parametric scattering in that two pump photons participate in the elementary act, is closely allied to this effect. We shall also look at some of the special features of the effect of light scattering by polaritons, which is midway between parametric scattering and Raman scattering by ion vibrations in the crystal lattice. These vibrations are accompanied by oscillations of the electromagnetic field inside the crystal. The polariton is the quantum of a macroscopic (averaged) field, i.e., a photon in a medium. Therefore, light scattering by polaritons, as well as three- and four-photon parametric scattering, should more properly be called light-by-light scattering in matter (the latter qualification

†To put it more precisely, it has a classical analog.
sets it apart from light-by-light scattering in a vacuum, which is an extremely weak—and as yet unobserved—effect of relativistic quantum electrodynamics.

Parametric scattering offers a unique opportunity to “prepare” two-photon states of the field and has a number of other interesting characteristics as well. The spectrum of scattered light is almost continuous throughout the range from radio frequencies to pump frequencies. Individual atoms do not produce radiation independently; rather, the entire sample produces radiation coherently, which results in a fairly sharp directivity of radiation forward along the pump beam. Photons of a single pair diverge at specified small angles to each other and to the pump beam, in accordance with the law of conservation of momentum upon the interaction of three photons.

The effects discussed in this book deserve further study as more than merely “exotic” manifestations of the quantum nature of light and the nonlinearity of matter. They have already found a number of useful applications. Parametric scattering forms the basis of a new spectroscopic method of measuring the linear and nonlinear optical parameters of crystals. Given powerful pulse pumping, it passes into a fairly intense parametric superluminescence, which provides a source of smoothly frequency-tunable short (up to $10^{-12}$ sec) light pulses. As the intensity of spontaneous three-photon parametric scattering is proportional to pump intensity, it can also be observed by means of nonlaser pumping sources—as distinct from four-photon scattering, which is proportional to the square of the pump, and parametric superluminescence, which, by definition, is superlinearly (exponentially) pump-dependent.

From the standpoint of applied nonlinear optics, the parametric scattering effect is the source of noise that limits the sensitivity of parametric amplifiers and light frequency converters and the ultimate stability of parametric light generators. However, quantum noise may, in principle, find useful applications in light metrology, where they can form the basis of “quantum photometry.” A parametric frequency converter can also serve as an absolute light brightness meter (one requiring no calibration). Besides, the simultaneous directed emission of photons in pairs in parametric scattering offers the opportunity of developing a practical standard photon generator that radiates a known number of photons. It can also be used to realize the famous gedanken experiment of Einstein, Podolsky, and Rosen.
Finally, the phenomena described in this book have a certain heuristic and pedagogical value, for they offer an opportunity to study, on the basis of a clear example, nonrelativistic quantum electrodynamics and also to master many important concepts of theoretical physics, such as Green functions, fluctuation-dissipation theorems, etc. And although the immediate subject matter of this book is fairly narrow, the author touches, in passing, upon a wide range of methodological questions of the theory of light-matter interaction. To give the reader a foretaste, here is a rundown of some of them, presented in a deliberately paradoxical form: Can an incandescent transparent material radiate light? Can one describe the Brown–Twiss effect using Kirchhoff’s law? Can a nonzero mean cube of an electric field exist in thermal radiation? Can one measure the brightness of electromagnetic vacuum fluctuations and design a light source with a known number of emitted photons? Do Stokes and anti-Stokes components correlate in the case of Raman scattering of light? Is it possible to create a field state with a definite energy and an indefinite number of photons? All these questions are answered in the affirmative in this book.

The author has drawn heavily on the results of his own work done as part of the research program maintained by the Department of Wave Processes at the Physics Department of Moscow State University. The author has been fortunate to work for many years under the expert guidance of the late Rem Khokhlov, an outstanding scientist and a charming person, who, together with S.A. Akhmanov, founded Moscow University’s school of nonlinear optics. Rem Khokhlov showed intense interest in the “photon” effects of nonlinear optics and the first rough draft of the present book had won his approval.

The author wishes to express his gratitude to the following: N.I. Nazarova, G.V. Venkin, V.S. Dneprovskii, D.P. Krindach, A.N. Penin, B.F. Polkovnikov, V.V. Fadeev, and other staff members of the Department of Wave Processes for their help and support. The author also acknowledges his indebtedness to P.V. Elyutin, B. Ya. Zel’dovich, Yu. A. Il’inškil’, and R.L. Stratonovich, who have read the manuscript and made valuable comments and suggestions. Finally, the author would like to share the responsibility for causing yet another book to see the light of day with V.B. Braginskii, at whose insistence he undertook the job of writing it.
List of Notation

A  cross-section of the sample or of the beam
\( A \)  absorption coefficient
e  unit polarization vector
E  electric field strength
\( \mathcal{E} \)  energy
F  photon flux density
\( \mathcal{F} \)  transformation coefficient
G  Green’s function
\( \mathcal{G} \)  amplification or transmission coefficient
H  magnetic field strength
\( \mathcal{H} \)  hamiltonian
\( j \)  current density
l  sample length
L  length of the quantization box
M  number of molecules
N  number of photons in one mode
\( \mathcal{N} \)  equilibrium number of photons per mode
P  polarization; probability
\( \mathcal{P} \)  power
S  brightness or intensity of light
S  scattering operator
\( u \)  group velocity
U  scattering matrix
\( \mathcal{U} \)  evolution operator
v  volume of one mode in k-space \( (v = 8\pi^3/L^3) \)
V  sample volume
\( \mathcal{V} \)  perturbation energy
W  photon flux
\( \Delta \)  phase velocity mismatching
LIST OF NOTATION

\( \eta \)  quantum efficiency
\( \theta \)  scattering angle
\( \nu \)  frequency in cm\(^{-1}\); polarization index
\( \pi \)  projection operator (on the plane, perpendicular to the wave vector)
\( \rho \)  angle between the group and phase velocities
\( \chi \)  nonlinear susceptibility (or polarizibility); characteristic function
\( \Omega \)  solid angle
1 Introduction

IN THIS CHAPTER, we present a preliminary qualitative and, it is hoped, clear description of the basic properties of parametric (three- and four-photon) and polariton light scattering (Sections 1.1–3). In addition, Section 1.4 looks at some basic concepts in statistical optics and Section 1.5 gives some background information on the discovery of these and other related multiphoton effects. A more detailed phenomenological treatment of scattering processes is given in Chapters 6 and 7.

1.1 Parametric Scattering

When a beam of light passes through transparent homogenous material—whether gas, pure liquid, or a perfect crystal—a small part of the light's energy is scattered in all directions by the atomic structure of the material. At low temperatures, ignoring quantum fluctuations, the atoms are fixed and light only changes its direction of propagation in scattering (elastic scattering), while, at high temperatures, the thermal motion of the atoms modulates the scattered light, changing not only the direction but also the frequency of the light (inelastic scattering). As a result, the frequency spectrum of scattered light repeats, subject to a shift into the optical region, the spectrum of the thermal motion of matter. For instance, in the case of ordinary Raman scattering, the spectrum of scattered light consists of several discrete components that are distant from the frequency of the incident light by an amount equal to the frequency of one of the normal vibrations of atoms in the molecule. As a rule, the frequency shift does not exceed a few percent in Raman scattering.
The Spectrum of Parametric Scattering

A characteristic feature of parametric scattering (PS) is the continuous spectrum of the scattered radiation, which, with short interruptions, may occupy the entire range from radio frequencies to the frequency of the incident light (pump), with the light of a given frequency $\omega_1$ being radiated by the material in a definite direction (Figure 1.1) that is dependent on the dispersion of the refractive index $n(\omega)$, according to the following equation:

$$k_1 + k_2 = k_3,$$

where $k_1$ is the wave vector of the observed scattered light and is equal in magnitude to $n_1 \omega_1/c = 2\pi n_1/\lambda_1$ ($\lambda_1$ is the wavelength in vacuum), $k_3$ is the wave vector of the pump, and $k_2$ is the wave vector of the idler wave, with a frequency of $\omega_2 = \omega_3 - \omega_1$. As the momentum of a photon in a medium is equal to $\hbar k$ ($\hbar$ is Planck's constant) (1), which relates to the case of three-photon parametric scattering, can be interpreted as the law of the conservation of momentum upon the interaction of three photons. It is also known as the phase-matching condition or the phase-velocity matching condition.

In the case of four-photon parametric scattering (or hyperparametric scattering, HPS) one has to add $k_3$, the wave vector of the second incident light beam, which may have a different direction and a different frequency, to the right-hand side of (1). This effect is interpreted as the result of the interaction of four photons. Its value is proportional to the product of the intensities of the two incident beams.

![Figure 1.1](image)

**Figure 1.1.** In parametric scattering of light, a pump photon with wave vector $k_3$ decays into a photon pair with wave vectors $k_1$ and $k_2$ under the laws of the conservation of energy and momentum. The resulting effect, which is visible to the naked eye, resembles a rainbow.
Thus, parametric scattering is attributable to the decay of one or two incident photons into a pair of photons with altered frequencies and directions. The sole restriction on the frequencies and directions, \((1)\), is fairly weak; this leads to a quasicontinuous parametric scattering spectrum.

The intensity of parametric scattering, like that of other multiphoton optical phenomena, can be determined by means of the nonlinearity of the reaction (response) of the material to the electric field of light:

\[
P(E) = \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \cdots,
\]

where \(P\) is the polarization, i.e., the dipole moment of the unit volume of the material. The linear polarizability \(\chi^{(1)}\) determines the index of refraction; the quadratic polarizability \(\chi^{(2)}\), which has an appreciable value only in piezocrystals, leads, in addition to three-photon parametric scattering, to light frequency doubling as well as other analogous nonlinear optical effects. The cubic polarizability \(\chi^{(3)}\) describes the tripling of the light frequency and the hyperparametric scattering. The same nonlinear parameters of matter determine the effect of the parametric amplification of light intensity, which has led to the development of parametric light generators, an important type of frequency-tunable lasers.

From the standpoint of radiophysics, scattered light is the inherent noise of a parametric amplifier. The specific character of the optical range manifests itself in the replacement of the noise temperature \(T\), which determines radio amplifier noise, by the energy of the quantum \(\hbar \omega\) divided by Boltzmann's constant \(\kappa\) and also in the "multimode" character of optical systems. This phenomenon is variously known as "parametric luminescence," "optical parametric noise," or "parametric splitting of the light frequency."

The effect of three-photon parametric scattering is literally observable. With the power of incident blue light at 0.1 \(\text{w}\), the green-yellow-red glow of a lithium niobate crystal is readily visible to the naked eye. The idler frequencies \(\omega_2\) lying in the infrared (IR) range correspond to the green glow in this case. As \(\omega_2\) approaches the natural frequencies of the crystal lattice, parametric scattering continuously changes into ordinary Raman scattering on the ion oscillations of the crystal lattice. The intermediate case has come to be known as "light scattering by polaritons."
Coherent Scattering

The condition (1) leads to the directivity of scattered waves of a given frequency, which always obtains if the individual radiating elements of matter are phase-matched and coherent. This spatial coherence is the result of the nonlocality of the radiating heterogeneity, i.e., of the propagation of the idler wave (Figure 1.2).

Coherence also characterizes other types of scattering, if they occur through collective excitations of matter with a nonzero propagation velocity. For instance, Mandelshtam-Brillouin scattering is the result of incident light scattering by sound waves. It also obeys (1) if it is understood that $k_2$ stands for the sound wave vector.

A characteristic feature of parametric scattering is the common nature of both waves—the scattering (idler) and observed (signal) waves—are equivalent. Besides, unlike sound waves, a light idler wave can propagate without damping within the material over a distance of 1 cm or more, i.e., it can go clean through a sample into the vacuum beyond its surface.

It should be emphasized that coherent scattering involves the addition of the amplitudes of the partial waves radiated by individual molecules of the sample. That is why the intensity of scattering at a given frequency and in a given direction is proportional to $M^2$, where $M$ is the number of molecules exposed to the light of the pump.

![Figure 1.2. Coherent light scattering. The light incident from the left is reflected from the wave fronts (dotted line) of the idler wave, giving rise to a signal (observed) wave of frequency $\omega_1 = \omega_3 - \omega_2$ and wave vector $k_3 = k_5 - k_1$. Idler waves exist in the sample because of thermal or quantum fluctuations and may be either sound or light waves. In the latter case, light-by-light scattering, otherwise known as parametric scattering, occurs.](image)
addition to coherent scattering, there is nondirectional, noncoherent scattering by individual molecules, which is proportional to $M$. We shall call this type of scattering, which turns the pump photon into a photon pair as a result of the interaction with individual molecules, three-photon Rayleigh scattering, as it is analogous to ordinary Rayleigh scattering, in which a pump photon is turned into a single photon with a different propagation direction. There also exists hyper-Rayleigh scattering, in which two pump photons are turned into a single photon with two frequencies.

**Zero Fluctuations of the Vacuum**

In the case of scattering by sound, optical heterogeneity results from the thermal motion of the material. But how do the scattering light waves responsible for parametric scattering get into a transparent crystal in the first place? One source is the thermal radiation of the surrounding walls, the ambient air, etc., which are nontransparent at the frequency $\omega_2$. The intensity of thermal radiation at the frequency $\omega$ at temperature $T$ is proportional to Planck’s function (Figure 1.3)

$$\mathcal{N}(x) = (e^x - 1)^{-1} \left( x = \frac{\hbar \omega}{\kappa T} \right),$$

where $C_2 = 2\pi \hbar c / \kappa = 1.4388 \text{ cm} \cdot \text{K}$, which is known as the second radiation constant. However, given $\lambda_2 = 0.5 \mu\text{m}$ and room temperature, $x \sim 100$ and $\mathcal{N} \sim 10^{-44}$; thus, it is necessary, in order to account for parametric scattering, to apply quantum radiation theory, according to which spontaneous effects can be calculated by adding unity to (3), which then means the mean number of photons in a single mode,

$$\mathcal{N}(x) \rightarrow \mathcal{N}(x) + 1 = -\mathcal{N}(-x).$$

To make this rule more vivid, we may assume that this unity is determined by the zero fluctuations of the vacuum that pervades the Universe. These quantum fluctuations of the idler field do not, however, make themselves felt in the case of anti-Stokes scattering ($\omega_2 > \omega_3$). A consistent description of the difference between Stokes and anti-Stokes scattering is only possible within the framework of the quantum theory of light.
6 INTRODUCTION

1.1

Figure 1.8. The Planck function determines the mean number of photons per mode of the equilibrium field in terms of the frequency of the mode $\omega$ and temperature.

The Effective Temperature of Parametric Scattering

Mandelstam–Brillouin scattering is caused by the modulation of the dielectric permeability $\epsilon = 1 + 4\pi\chi^{(1)}$ by a thermal sound wave having an amplitude of $Q$, the elasto-optic constant $\partial\chi^{(1)}/\partial Q$ having been taken into account. In parametric scattering, the role of this constant, which violates the principle of light-wave superposition in matter, is played by the quadratic polarizability $\chi^{(2)} \approx \partial\chi^{(1)}/\partial E_2$. Let the amplitudes of the electric field in the pump wave and in the idler wave equal $E_3$ and $E_2$; then, at point $r$ in the material, polarization will occur at the observed frequency

$$P_1(r) = \chi^{(2)} E_3 E_2^* e^{i(k_3 - k_2) \cdot r}. \quad (5)$$

Strictly speaking, the polarizability $\chi^{(2)}$ is a third-rank tensor (as it links three vectors); however, we shall ignore the vectorial nature of the electromagnetic field in this chapter. We shall, however, note that tensors of the third rank only have appreciable values in noncen-
trosymmetric media. Piezocrystals have this property in the absence of a static external force, which explains why three-photon parametric scattering has only been observed in them thus far.

It follows from Maxwell's equations that a polarization of amplitude $P_1$ oscillating with a frequency $\omega_1$ radiates a wave with the amplitude

$$E_1 = 2\pi i\omega_1 c^{-1} \int_{-l/2}^{l/2} dz P_1 e^{i(p_x - k_{1z})z},$$

where $l$ is the sample thickness (which, we assume, has the shape of a planar layer) and $p$ is the wave vector of polarization, which, according to (5), equals $k_3 - k_2$ (in this chapter, we neglect the coefficients of the order of unity). It follows from (6) that the signal has a maximum amplitude if the phase-matching condition is satisfied (1) for longitudinal components of wave vectors (in the case of a layer, the phase-matching condition for transverse components of wave vectors must be rigorously satisfied), with the field being proportional to $l$:

$$E_1 = 2\pi i\omega_1 c^{-1} P_1 l.$$  

(7)

In the case of inexact phase-matching of longitudinal components, one must replace $l$ by the "length of coherence":

$$l_\Delta \equiv \int_{-l/2}^{l/2} dz e^{-iz\Delta} = \frac{\sin(l\Delta/2)}{\Delta/2},$$

(8)

$$\Delta \equiv \Delta(k_1) \equiv k_{1z} + k_{2z} - k_{3z},$$

(9)

With the given vector $k_1, k_3$, the condition $\omega_2 = \omega_3 - \omega_1$, and transverse phase-matching, the idle wave vector $k_2$ is determined to within the sign of its projection on the $z$ axis.

In order to use the quantization rule (4), it is necessary to turn from field amplitudes to the number of photons per mode, $N \sim |E|^2/\hbar \omega$. As a result, we find the number of photons per mode to the right of the layer sample from (5-8):

$$N'_1 = \mathcal{F}(k_1)(N_2 + 1),$$

(10)
where \( N_2 \) is the number of real idler photons incident on the sample from the left in the direction \( \mathbf{k}_2 \). When \( N_2 \gg 1 \), this formula describes the stimulated effect of frequency subtraction. The function \( \mathcal{F}(k) \) determines the energy conversion coefficient of idler waves into signal waves. Let \( \lambda_1 = \lambda_2 = 2\lambda_3 = 0.5 \text{ \mu m} \), \( \chi = 10^{-8} \text{ cgs units} \), \( \Delta = 0 \), \( l = 1 \text{ cm} \), and the pump intensity

\[
S_3 = \frac{c}{2\pi} |E_3|^2
\]

be equal to \( 1 \text{ w} \cdot \text{cm}^{-2} \), then

\[
\mathcal{F} = \frac{(2\pi)^6}{c} \left( \frac{\chi l}{\lambda_1} \right)^2 S_3 = 10^{-7}.
\]

This estimate shows that, in the case of three-photon processes, effective frequency transformation in piezocrystals is only possible if the pump intensity is on the order of hundreds of megawatts per \( \text{cm}^2 \), something achieved only in pulsed lasers.

With \( N_2 = 0 \) (i.e., with no real idler wave present at the "input"), (10) describes parametric scattering. In this case, according to the estimate (12), the number of scattered photons per mode equals \( N'_1 = 10^{-7} \). Let us determine, by means of (3), the effective or brightness temperature of the light scattered in the direction \( \mathbf{k}_1 \) as follows:

\[
T_{\text{eff}}(\mathbf{k}_1) \equiv \frac{\hbar \omega_1}{\kappa \ln(1 + 1/N'_1)} = \frac{C_2}{\lambda_1 x_{\text{eff}}}.
\]

Thus, the effective temperature is the temperature of an equilibrium Planck field that has the same number of photons per mode with a frequency \( \omega_1 \). It follows from (12) and (13) that, in the phase-matching direction \( x_{\text{eff}} = 16 \) and \( T_{\text{eff}} = 1800 \text{ K} \), which corresponds to a quantity readily visible to the naked eye.
The Number of Photons per Mode and the Brightness of Light

How are the number of photons in a mode or effective temperature connected with directly observable quantities, e.g., with luminosity or the number of pulses per second $W$ at the output of a photomultiplier tube? In the absence of an actual resonance cavity, the notion of a mode can be introduced by means of the mental isolation in space of a parallelepiped with sides $L_x, L_y,$ and $L_z$ and volume $L^3$ that covers the area of the field of interest. Furthermore, the real spatial distribution of the field $E(x)$ (at a fixed instant of time) is replaced by the periodic function $E_{\text{per}}(x)$, which has a period $L_x$ along the $x$ axis, $L_y$ along $y$, and $L_z$ along $z$. Inside $L^3$, the field $E_{\text{per}} = E$, while the difference between the real and periodic fields outside $L^3$ is of no importance (Figure 1.4).

The periodic function of $x$ can be expanded into a Fourier series (it contains only those harmonics which are a multiple of the quantity $2\pi/L_x$); hence, the periodic field can be represented in the form of a triple sum:

$$E(x) = \sum_k E_k e^{i k \cdot r},$$  \hfill (14)

where we introduced a wave vector

$$k = \left\{ \frac{2\pi m_x}{L_x}, \frac{2\pi m_y}{L_y}, \frac{2\pi m_z}{L_z} \right\},$$  \hfill (15)

$$m_\alpha = 0, \pm 1, \pm 2, \ldots$$

Thus, the field distribution in space is now determined not by a continuous set $E(x)$, but rather by a countable set $E_k$, which is numbered by the integers $m_\alpha$. The vectors $k$ form the $k$ space, which is split by

![Figure 1.4. On the definition of quantization length. $E_x(z)$ is the real field; $\tilde{E}_x(z)$ is the fictitious field periodic in space.](image-url)
the periodicity condition (15) into cells or modes with the "volume" 
$(2\pi)^3/L_xL_yL_z \equiv v$.

Let us now allow for the dependence of a field on time. It follows from Maxwell's equations (Section 3.1) that, if $L^3$ lacks "sources" or "drains" of the field, then

$$E_k(t) = E_k^{(+)} e^{-i\omega_k t} + E_k^{(-)} e^{i\omega_k t}, \quad (16)$$

where

$$\omega_k \equiv ck = 2\pi c \left[ \left( \frac{m_z}{L_z} \right)^2 + \left( \frac{m_y}{L_y} \right)^2 + \left( \frac{m_x}{L_x} \right)^2 \right]^{1/2}, \quad (17)$$

is the mode's natural frequency. The complex numbers $E_k^{(\pm)}$ are the amplitudes of the field in a mode.

Let a detector "see" waves with a single polarization belonging to a certain volume in the $k$ space containing many modes,

$$\Delta^3 k = k^2 \Delta k \Delta \Omega = \omega^2 \Delta \omega \Delta \Omega / c^3 \gg v, \quad (18)$$

where $\Delta \omega$ is the detector's frequency band and $\Delta \Omega = \sin \theta \Delta \theta \Delta \phi$ is its angular aperture. Let us introduce the notation

$$\Delta g \equiv \Delta^3 k / v \equiv g_{\omega \Omega} \Delta \omega \Delta \Omega \quad (19)$$

for the number of modes "seen" by the detector ($g_{\omega \Omega} = \omega^2 / c^3 v$ is the mode density).

Let each mode contain $N$ photons; the photon density in the mode is then $NL^{-3}$, the photon flux density in the mode is $cNL^{-3} \equiv F$, and the photon counting rate is

$$W = \eta FA \cos \theta \Delta g, \quad (20)$$

where $\eta$ is the quantum efficiency of the photomultiplier tube, $A$ is its area, and $\theta$ is the angle of photon incidence on the photomultiplier tube (PMT) surface. If the observed modes contain unequal numbers of photons and if the quantum efficiency is likewise nonuniform, then,
obviously, we should write

\[ W = \sum_k \int dA \eta_k F_k \cos \theta_k, \tag{21} \]

instead of (20). Let \( L^3 \) tend to infinity so that we may turn from summation to integration according to the rule

\[ \lim_{v \to 0} \sum_k f_k = v^{-1} \int dk f(k) = \frac{\omega^2}{vc^2} \int_0^\infty d\omega \int_{4\pi} d\Omega f(\omega, \Omega); \tag{22} \]

as a result, (21) takes on the form \((A_\perp \equiv A \cos \theta)\)

\[ W = \int dA_\perp \int d\omega \int d\Omega \omega^2 N/8\pi^3 c^2 \]

\[ = \int dA_\perp d\omega d\Omega \eta S_{\omega \Omega}/k\omega, \tag{23} \]

where we have introduced a new quantity: the spectral brightness of radiation, \( S_{\omega \Omega} \), which is equal to the energy transferred across a unit of the detector surface per unit time in unit spectral interval and solid angle. This quantity is often called, simply, intensity, its dimensions being erg \( \cdot \) sec\(^{-1} \) \( \cdot \) cm\(^{-2} \) \( \cdot \) steradian\(^{-1} \) \( \cdot \) Hz\(^{-1} \).

In an absorbing, radiating, or scattering medium, brightness and the number of photons also depend on the spatial coordinate: \( S_{\omega \Omega} \equiv S(k, r) \), where \( r \) is the coordinate of the center of the quantization box. Here the coordinate along the \( x \) axis is determined only to within the "Fourier uncertainty,"

\[ \delta x \sim \pm \frac{L_x}{2} \equiv \pm \frac{\pi}{\delta k_x}, \tag{24} \]

where \( \delta k_x \) is the interval between the neighboring modes, i.e., the function \( S(k, x) \) is constant within the limits of a single cell with the volume \( vL^3 = 8\pi^3 \) in the six-dimensional phase space \( r \times k \). If no absorption or scattering occurs in the path \( l \) along the radiation direction \( k \) then

\[ S(k, x) = S(k, x + l\hat{k}), \tag{25} \]
§1.1

— the brightness of light in the given direction does not diminish with increasing distance from the light source in this direction ($\kappa = k/k$).

$S$ is also time-dependent in the nonstationary case.

In photometry, brightness is commonly considered to be a property of the light source and not of space, i.e., a function of the coordinate $r_A$ belonging to the surface of a continuous body. On the other hand, when the transfer of light energy in a turbulent medium is considered, the notion of light intensity at an arbitrary point in space $r$ is used. These concepts are essentially synonymous ("surface brightness" is transferred to any point of the empty space according to (25)) and we shall speak of "the brightness of light at point $r$." We shall define brightness in rigorous statistical terms later (see (3.2.8.)). We shall refer to the quantity $S = h\omega cN/L^3$, which characterizes the density of the energy flux in a single mode, as intensity.

Thus, according to (19) and (23), the spectral brightness of light and the number of photons in a mode are connected by the relationship

$$S_{\omega \Omega} = e^{\hbar \omega g_{\omega \Omega} N/L^3} = NS_{\omega \Omega}^{\text{vac}} \quad (S_{\omega \Omega}^{\text{vac}} \equiv h\omega^3/8\pi^3c^3). \quad (26)$$

Substituting the Planck distribution (3) for $N$, we obtain the thermal radiation brightness (of a single polarization) of an ideal black body. The proportional constant in (26) may be called the spectral brightness of vacuum fluctuations. Equilibrium radiation will have this kind of brightness if unity is substituted for $N$ in Planck's formula. Let us find the effective "vacuum temperature," i.e., the temperature of equilibrium radiation that has one photon per mode (at a given frequency). By definition, $N(T_{\text{vac}}) = 1$, and it follows from (3)—given $\lambda = 1 \mu m$—that $T_{\text{vac}} = C_2/\lambda \ln 2 \sim 2 \times 10^4$ K.

It is sometimes convenient to refer radiation energy to a unit interval of the wavelength. The following relationship obviously holds:

$$S_{\lambda \Omega} = \left| \frac{d\omega}{d\lambda} \right| S_{\omega \Omega} = \frac{2\pi c}{\lambda^2} S_{\omega \Omega}. \quad (27)$$

According to (26) and (27), "the spectral brightness of a vacuum" with $\lambda = 1 \mu m$ equals

$$S_{\lambda \Omega}^{\text{vac}} = \frac{C_1}{2\pi \lambda^5} = 0.5955 \text{ w} \cdot \text{A}^{-1} \cdot \text{cm}^{-2} \cdot \text{steradian}^{-1}, \quad (28)$$
§1.1 PARAMETRIC SCATTERING

where \( C_1 \equiv 4\pi^2\hbar c^2 \) is the first radiation constant.

Integral quantities with less detailed information are also used in photometry. If the radiation spectrum is narrower than the pass band of the detector, then it measures

\[
S_\Omega = \int d\omega S_{\omega \Omega}. \tag{29}
\]

This quantity is called brightness. The integral

\[
S_\omega = \int d\Omega \cos \theta S_{\omega \Omega} \tag{30}
\]

might be called spectral illumination intensity (or spectral luminosity), while the integral

\[
S = \int d\omega d\Omega \cos \theta S_{\omega \Omega} \tag{31}
\]

might be called luminous radiance. Finally, if the detector intercepts all the radiation, then the power (or luminosity) of the source is measured:

\[
P = \int d\omega d\Omega dA \left( S_{\omega \Omega} \right). \tag{32}
\]

It is sometimes convenient to use the following notation:

\[
S_{\omega \Omega} = \frac{dS}{d\omega d\Omega} = \frac{dP}{d\omega d\Omega dA} = \frac{d\mathcal{E}}{d\omega d\Omega dA dt}, \tag{33}
\]

where \( \mathcal{E} \) is the total energy transferred across a particular plane.

The Brightness of Parametric Scattering

According to (10), (11), and (26),

\[
S_{\omega \Omega} = S_{\omega \Omega}^{\text{vac}} \mathcal{F} = \hbar c^{-5} \omega_1^4 \omega_2 \chi^2 S_{3/\Delta}^2. \tag{34}
\]
The power scattered in unit spectral and angular intervals is \( A_3 \cos \theta_1 \) times greater than \( S_{\omega \Omega} \) and is proportional to the pump power:

\[
P_{\omega \Omega} \approx \hbar c^2 \omega_1^2 \omega_2 \chi^2 i_{\Delta}^2 P_3,
\]

(35)

where \( P_3 = S_3 A_3 \) is the power and \( A_3 \) is the cross-section of the pump beam \((\theta_1 \approx 0)\). From this we find that the differential probability of the transformation of the pump photon with frequency \( \omega_1 \) and direction \( \hat{k}_1 \), within the limits of unit intervals with \( \theta_1 \ll 1 \), equals

\[
P_{\omega \Omega} = \frac{\omega_2 P_{\omega \Omega}}{\omega_1 P_3} = \hbar c^2 \omega_1^2 \omega_2 \omega_3 \chi^2 i_{\Delta}^2,
\]

(36)

\[
P_{\lambda \Omega} = \frac{(2\pi)^6 \hbar c^2 \chi^2 i_{\Delta}^2}{\lambda_1 \lambda_2 \lambda_3} \sim 4 \times 10^{-7} \text{nm}^{-1} \cdot \text{steradian}^{-1}.
\]

The latter estimate is made for the same conditions as in (12). Let us also introduce the differential flux of signal photons with a dimension of steradian\(^{-1}\):

\[
W_{\omega \Omega} \equiv \frac{P_{\omega \Omega}}{\hbar \omega_1} = e^{-\delta} \omega_1^2 \omega_2 \chi^2 i_{\Delta}^2 P_3.
\]

(37)

**Parametric Superluminescence**

We have not yet allowed for the back action of the signal wave on the idler wave. The relationships (5) and (6), albeit with interchanged indices 1 and 2, also hold for this case. Together, these relationships form a system of linear differential equations that describes the mutual amplification of the signal and idler waves as they pass through a nonlinear layer. This system is easily solved with a transformation coefficient equal to (Figure 1.5)

\[
F_k = \beta^2 \frac{\sinh^2 \sqrt{\beta^2 - \Delta^2/4}}{\beta^2 - \Delta^2/4},
\]

(38)

where the amplification index \( \beta \) is defined in (10). If weak noncoherent radiation with \( N_k \) photons in modes is incident on the layer, in
addition to the pump photons, then the layer will radiate $N_k'$ photons, where

$$N_k' = G_k N_k + \mathcal{F}_k (N_k + 1)$$

(39)

$$= G_k \left( N_k + \frac{1}{2} \right) + \mathcal{F}_k \left( N_k + \frac{1}{2} \right) - \frac{1}{2},$$

(40)

$$G_k = \mathcal{F}_k + 1$$

(41)

($\vec{k} \perp \equiv -\vec{k} \perp$ and $\omega_\perp = \omega_3 - \omega_k$). It follows from (39) that spontaneous and stimulated effects are closely related. Both are described by a single formula. We will refer to formulas of this type as generalized Kirchhoff's laws.

Note that, by virtue of (41), the intensity of spontaneous radiation may be calculated by means of classical formulas by the addition of $\alpha$ photons to the input of the signal channel and $1 - \alpha$ at the input of the idler channel and by subtracting $\alpha$ photons from the number of signal photons at the output. For instance, we obtain a symmetric formula (40) with $\alpha = 1/2$. This rule is a generalization of (4) that is only valid for the Stokes frequencies.

Let the incident field be absent ($N_i = 0$; there is a vacuum at the input); then, according to (39), each output mode will have $\mathcal{F}_k$ photons. The radiation will reach its maximum in the directions and at the frequencies that satisfy the phase-matching condition (1), and, provided that the pump power or the sample's thickness is sufficient (so that $\beta l \gg 1$), $N_k'$ will be exponentially dependent on $E_3$ and $l$:

$$N_k' = \sinh^2 \beta l \approx e^{2\beta l/4}.$$  

(42)

This phenomenon has come to be known as parametric superluminescence (by analogy with superluminescence—enhanced spontaneous radiation in optical quantum amplifiers with population inversion).

Equations (13) and (34) for the effective temperature and radiation brightness are also valid in the case of superluminescence if we use a corrected transformation coefficient (38) in them (or replace $\Delta/2$ in (8) by $\sqrt{\Delta^2/4 - \beta^2}$ and use the formula $\sin i x = i \sinh x$). In this case if, for instance, $\mathcal{F} = 10$, then $T_{\text{eff}} \approx \hbar \omega \mathcal{F}/k \sim 10^6$ K.

It should be noted that a model of a plane sample is meaningful for describing superluminescence only within the narrow cone of those directions along the pump beam which satisfy the condition
Figure 1.5. The intensity of parametric superluminescence as a function of frequency with different pump intensities.

$\theta_i \ll a/l$ ($\theta_i$ are the scattering angles for the signal and idler waves, i.e., the angles between $k_i$ and $k_3$, and $a$ is the radius of the pump beam). If we create feedback (by means of mirrors, say), then we would convert the amplifier into a parametric light generator. Zero fluctuations in the process act as a "trigger" that sets off self-excited oscillations.
Tuning Curves

The proportionality of the parametric scattering intensity (of given frequency and direction) to the “phase-matching function” $l_\Delta$ (see (8)) determines a fairly unusual conic structure of the scattered field, one reminiscent of Cherenkov radiation. The phase-matching condition $\Delta = 0$, which may be interpreted as a consequence of the laws of the conservation of energy and momentum during the interaction of three photons, defines the “phase-matching surface” on which the radiation reaches its maximum in $k_1$ space (i.e., in a space formed by the possible wave vectors of the signal).

Phase-matching is possible in optically isotropic media if there is an area with an anomalous dispersion of the refraction index ($dn/d\omega < 0$) between the frequencies $\omega_1$ and $\omega_2$, since, otherwise, because of normal dispersion, $k_1 + k_2 < k_3$. In particular, anomalous dispersion can be caused by a resonance of the light frequency with the natural oscillations of the crystal lattice. The frequency of these oscillations $\nu_\mu = \omega_\mu/2\pi c$, lies in the infrared range and is on the order of hundreds and even several thousands of cm$^{-1}$. The pump frequency usually corresponds to the visible region of the spectrum ($\nu_3 \sim 2 \times 10^4$ cm$^{-1}$, $\lambda_3 = \nu_3^{-1} \sim 0.5 \mu$m); thus, in isotropic crystals, phase-matching occurs only in a very narrow interval of frequencies near the pump frequency. The resonance of the idler frequency with the lattice frequencies leads not only to an increased $k_2$, but also to light absorption on the length $l/\alpha_2$, which is far smaller than the scattering region $l$. As a result, practically no idler photons are radiated. We classify this case as belonging to Raman scattering by polaritons rather than to parametric scattering (for details, see Section 1.2 and Chapter 6).

However, in birefringent crystals, phase-matching is also possible when all three frequencies belong to one and the same “window” of transparency sandwiched between lattice and electronic ($\nu_\mu \sim 10^5$ cm$^{-1}$) frequencies. In such crystals, monochromatic incident light excites two waves differing in their propagation velocities $c/\tau$ (i.e., wavelength $\pi/\nu = 2\pi/k$) and in the polarization directions of their electric fields $e$ with respect to crystallographic axes of the crystal.

In the case of uniaxial crystals, a wave with an $eE$ field perpendicular to the axis of the crystal $c$ is called ordinary ($n = n^0$), while
the second, independent, wave, in which e and c are coplanar, is called extraordinary \((n = n^e)\). In most piezocrystals used in applied nonlinear optics, \(n^0(\omega) > n^e(\omega)\) and two types of phase-matching are possible: \(k^0_1 + k^0_2 = k^3_3\) and \(k^0_1 + k^2_2 = k^3_3\). In the former case, the phase-matching surface has a symmetry axis along \(k_3\) and determines the tuning curve \(\omega_1(\theta_1)\), where \(\theta_1\) is the scattering angle, i.e., the angle between \(k_1\) and \(k_3\). Figure 1.6 shows the dependencies \(\lambda_1(\theta_1)\) for lithium niobate with \(k_3 \perp c\) for different pump wavelengths. It follows from the plots that, given \(\lambda_3 \lesssim 0.53\ \mu\text{m}\), the collinear phase-matching \((\theta_1 = \theta_2 = 0)\) necessary for superluminescence is possible. If the directions \(k_3\) and \(c\) form an angle \(\alpha_3\) that is less than 90°, then collinear phase-matching occurs at a larger pump wavelength.

As \(k_1\) moves away from the phase-matching surface, the function \(l_\Delta\) rapidly diminishes, obeying the law \((\sin x)/x = \text{sinc} x\), where \(x = l_\Delta/2\) and \(\Delta = \Delta(\omega_1)\). Let us find the frequency width \(\Delta \omega\) of the tuning curve with the change in \(\omega_1\) from the condition \(l_\Delta = 2\pi\).

![Figure 1.6. The relation between the scattering angle and the wavelength of parametric scattering with different pump wavelengths in the case of lithium niobate.](image)
dependence $\Delta(\omega_1)$ may, as a rule, be regarded as linear:

$$\Delta(\omega_1) = \frac{\partial \Delta}{\partial \omega_1} (\omega_1 - \omega_1^0),$$

$$\frac{\partial \Delta}{\partial \omega_1} = \frac{\partial k_{1x}}{\partial \omega_1} + \frac{\partial k_{2x}}{\partial \omega_1} \approx \frac{1}{u_1} - \frac{1}{u_2},$$

(43)

where $u = d\omega/dk$ is the group velocity and we have assumed that $\theta_i \ll 1$. Hence,

$$\Delta \omega_1 = \frac{2\pi}{l} \left| \frac{\partial \Delta}{\partial \omega_1} \right|^{-1} \approx \frac{2\pi}{|\tau_1 - \tau_2|} \left( \frac{l}{u_i} \right).$$

(44)

Thus, the spectrum width radiated in the given direction is determined by the mutual delay $\tau_1 - \tau_2$ of the idler and signal waves on the interaction length $l$. The angular width of the tuning curve is determined analogously:

$$\Delta \theta_1 = \frac{2\pi}{l} \left| \frac{\partial \Delta}{\partial \theta_1} \right|^{-1} \approx \frac{\lambda_3}{l \theta_2} \left( \theta_i \ll 1 \right).$$

(45)

In lithium niobate with $\lambda_3 = 0.48 \mu m$, $\lambda_1 = 0.67 \mu m$, and $l = 1 \text{ cm}$, the widths have the following orders of magnitude: $\Delta \nu_1 \sim 8 \text{ cm}^{-1}$ and $\Delta \theta_1 \sim 0.5'$. Integral Intensities of Parametric Scattering

The product of the effective spectrum width (44) and the double differential probability of scattering (36) with $\Delta = 0$ gives a scattering probability in a unit solid angle in a given direction:

$$P_\Omega = \frac{2\pi \hbar \omega_1^2 \omega_2 \omega_3 \chi^2 l}{c^5 |u_1^{-1} - u_2^{-1}|} \sim 3 \times 10^{-7} \text{ steradian}^{-1}.$$  

(46)

In ordinary Raman scattering in condensed media, the probability of the decay of a pump photon into a Stokes photon and a phonon has the same order of magnitude. It will be noted that the quantities $\omega_{1,2}$ and $u_{1,2}$, as well as $\chi$, in (46) are functions of the scattering angle $\theta_1$.
because of the $\Delta = 0$ connection. In certain "degenerate" directions, $u_1 = u_2$ and $P_\Omega$ grows dramatically. In these directions, the steepness of the tuning curve $|d\omega_1/d\theta_1| \to \infty$ and the linear approximation of the function $\Delta(\omega_1)$ is insufficient.

It is clear from Figure 1.6 that all the scattered radiation in the visible region is concentrated in a fairly narrow cone with a divergence angle on the order of a few degrees (this is the result of the small difference between $n^0$ and $n^s$). The energy radiated within this cone at the given frequency $\omega_1$ with $\theta_i \neq 0$ is proportional to the integral

$$
\int_0^{2\pi} d\phi_1 \int_0^\pi d\theta_1 \sin \theta_1 l^2 \Delta \approx \frac{4\pi^2 l k_2}{k_1 k_3}
$$

(47)

(we have used (45) and the relation $k_1 \sin \theta_1 = k_2 \sin \theta_2$, which follows from the condition of transverse phase-matching). From (47) and (36), we find the scattering probability in a unit spectral interval (irrespective of direction):

$$
P_\omega = h l (2\pi \omega_1 \omega_2 \chi / c^2)^2,
$$

$$
P_\lambda = (2\pi)^7 h c \chi^2 l \lambda_1^{-4} \lambda_2^{-2} \sim 10^{-10} \text{ nm}^{-1}.
$$

(48)

Hence, given a pump power of $P_3 = 1 \text{ w} (W_3 = 3 \times 10^{18} \text{ sec}^{-1})$, the spectral power of the signal is

$$
P_\lambda = \lambda_3 P_\lambda P_3 / \lambda_1 \sim 5 \times 10^{-11} \text{ w} \cdot \text{ nm}^{-1}.
$$

(49)

The full probability of the decay of the pump photon on the path $l$ (given the presence of phase-matching throughout the frequency interval $0-\omega_3$) will be on the order of

$$
P = \int_0^{\omega_3} d\omega_1 P_\omega \sim h c^{-4} \omega_3^5 \chi^2 l \sim 10^{-7}.
$$

(50)

The idealized model used here presupposes a transparent and optically homogeneous crystal of infinite cross section and a monochromatic planar pump wave. When these conditions are violated, (8),
which describes the detailed frequency-angular shape of the spectrum, will not be correct. For instance, if the pumping divergence $\Delta \theta_3$ is considerably greater than $\Delta \theta_1$ (see (45)), then the angular radiation width with a given frequency will have the order $\Delta \theta_3$, while the radiation intensity in the direction of phase-matching will be proportional not to $l^2$ but to $l_{\text{coh}}$, where the coherence length will be on the order of

$$l_{\text{coh}} \sim l \frac{\Delta \theta_1}{\Delta \theta_3} \sim \frac{\lambda_3}{\theta_2 \Delta \theta_3}. \quad (51)$$

Similarly, the crystal’s opacity at the idler frequency leads to the replacement of $l$ in (44) and (45) by the range of the idler photon: $l_{\text{coh}} \sim 1/\alpha_2$, where $\alpha_2$ is the absorption coefficient. In this case, usually, $u_2 \ll u_1$; thus, it follows from (44), with $l$ replaced by $l_{\text{coh}}$, that $\Delta \omega_1 \sim 1/\alpha_2 u_2 = 1/r$, where $r$ is the lifetime of the polariton.

It is significant that a decrease in the coherence length does not change the area of the spectral line, $\int d\omega l_\Delta^2$ or $\int \omega d\omega l_\Delta^2$, which is proportional to the crystal length; for this reason, the integral relationships (46), (48), and (49) remain valid.

### 1.2 Scattering by Polaritons

#### Polaritons and the Dispersion Law

As the frequency of the signal $\omega_1$ approaches the pump frequency $\omega_3$, the idler frequency $\omega_2$ approaches the natural frequencies of the crystal lattice $\omega_\mu$, which lie within the range from $10^2$ to $10^3$ cm$^{-1}$. In this process, parametric scattering continuously develops into scattering by polaritons (SP), while, in the case of an exact coincidence of $\omega_2$ with one of the frequencies $\omega_\mu$, it changes into Raman scattering (RS) by optical phonons. A polariton is a quantum of mixed electromagnetic and mechanical waves that arises in ionic crystals when infrared light is incident upon them. Mixed excitations in the region of electronic frequencies are also often called “polaritons.” The electric field of a light wave with $\omega \sim \omega_\mu$ causes the charged ions of the lattice to oscillate and thus irradiate light. As a result, the
group velocity of the wave $u$ drops sharply (almost to zero at an exact resonance), while its phase velocity becomes a nonmonotonic function of the frequency (the so-called anomalous dispersion). The functions $u(\omega) = d\omega/dk$ and $n(\omega) = kc/\omega$ are unambiguously associated with the law of polariton dispersion, i.e., the dependence $\omega(k)$ (Figure 1.7). Under the law of the conservation of momentum during scattering, the link $\omega_2(k_2)$ determines the observed tuning curve $\omega_1(\theta_1)$ by trigonometric formulas.

Thus, scattering by polaritons makes it possible to study lattice dynamics in an area of the $k$ space (the center of Brillouin’s zone) difficult to reach by other methods of investigation.

Figure 1.7. The dispersion of light $k(\omega_2)$ in the IR region (solid lines in the left-hand plot) and the function $k(\omega_2, \theta_1) \equiv |k_3 - k(\omega_3 - \omega_2)|$ (dotted lines). The section points of the plots $k$ and $k$ correspond to the phase-matching condition, i.e., they define the observed dependence between the scattering angle and the frequency $\theta_1(\omega_1)$ (right-hand plot).
Comparison of Parametric Scattering and Scattering by Polaritons

But what is the difference between scattering by polaritons and parametric scattering (i.e., scattering by photons in a medium)? After all, the concepts of polaritons and photons in a medium are essentially identical.

We can formally distinguish between them if we assume, for instance, that, in the case of photons, more than 50 percent of the energy of excitation is contained in the transverse electromagnetic field. Or, since it is experimentally more convenient to measure group velocity, we may draw the line between them with the retardation condition \( c/u = n = \omega dn/d\omega \), by, say, three or ten times. Besides, scattering by photons and polaritons may be distinguished by the mechanism of nonlinear polarizability. In the region of lattice frequencies \( \omega_\mu \) a resonant part \( \chi^{(2)}_{\text{res}} \) is added to the purely electronic nonlinearity \( \chi^{(2)}_\infty \); it is associated with the interaction between the vibrations of the atoms as a whole and the movement of electrons (this nonlinearity also determines ordinary Raman scattering at large angles at the frequencies \( \omega_1 = \omega_3 - \omega_\mu \)). Finally, "parametric scattering" is often taken to mean scattering by polaritons of the "upper" branch (see Figure 1.7).

However, we believe it advisable to retain a special term for parametric scattering on the strength of the transparency criterion of the sample for the idler wave: \( \alpha_2 \ll 1 \) (the conditions \( \alpha_{1,3} \ll 1 \) are, as a rule, satisfied in experiment). In this case, the signal and idler waves are equivalent; they go clean through the sample without attenuation, i.e., they are the total excitations of the entire space both inside and outside the sample. As a result, the intensity of parametric scattering, as distinct from scattering by polaritons, should not be directly dependent (via Planck's function (1.1.3)) on the sample temperature even when \( \hbar \omega_2 \ll \kappa T \). The transparency criterion is also convenient for distinguishing between four-photon parametric scattering (hyperparametric scattering) and hyper-Raman scattering by polaritons (Section 1.3). According to our definition, the borderline between parametric scattering and scattering by polaritons depends on experimental conditions—on the dimensions of the sample and on the absorption coefficient, which is, in turn, temperature dependent.

The proposed method of distinguishing between parametric scattering and scattering by polaritons is justified by the extraordinary
statistical properties of light radiated by a sample during parametric scattering. As mentioned in the introduction, correlated (by direction, frequency, and radiation time) pairs of signal and idler photons ("biphotons") may be of interest in applied optics for the development of a standard photon generator and for the absolute calibration of photomultiplier tubes (Section 6.4). The possibility of an absolute measurement of light brightness when comparing transformed frequency radiation with scattered radiation is also attributable to the transparency of the sample at the idler frequency.

The Intensity of Scattering by Polaritons

In accordance with our definition, incident idler radiation from outside does not penetrate deep into the crystal during scattering by polaritons because of the severe absorption in the infrared region. However, when $\alpha_2 l \gg 1$, there is Planck equilibrium radiation associated with thermal ion vibrations inside the crystal. The field $E_2$ "beats" with the pump field $E_3$ because of the quadratic nonlinearity $\chi^{(2)}$ and sets up the polarization (see (1.1.5)), which is the source of the observed scattering field. In the Stokes case, unity is added to the equilibrium number of photons $\mathcal{N}_2$, which allows for the fluctuations of the crystalline "polariton vacuum." Thus, scattering by polaritons is caused by the fluctuations of the idler field inside the scattering substance, while parametric scattering is due to external causes. In the intermediate case of a semitransparent sample, scattering intensity depends on two temperatures—the temperature of the sample and that of the incident field.

It will be convenient to describe scattering by polaritons phenomenologically if we introduce an increment of the dielectric permeability $\Delta \varepsilon$ at the frequency of the signal that appears upon switching on the pump. It can easily be shown (Section 6.6) that, given $\alpha_2 l \gg 1$, $\Delta \varepsilon$ has the following form:

$$\Delta \varepsilon(\omega_1, k_1) = 4\pi |E_3|^2 \left[ \chi^{(3)} + \frac{4\pi \chi^{(2)}}{n_2^2 - \varepsilon^*(\omega_2)} \right], \quad (1)$$

where $n_2 \equiv c|k_3 - k_1|/\omega_2$. Using this parameter, one may express both the stimulated effect of the parametric amplification of the signal
and the intensity of scattering by polaritons. In Section 6.6, we shall show that the scattering coefficient (see (1.1.37)) equals

\[ W_{\omega \Omega}(k_1) = -\Delta \varepsilon''_1(N_2 + 1)V/\lambda_1^3, \tag{2} \]

where \( V \) is the scattering volume and the imaginary part of \( \Delta \varepsilon_1 \) is negative, which corresponds to the effect of parametric and Raman amplification in the Stokes region.

**The Frequency-Angular Spectrum of Scattering by Polaritons**

In the resonance region, the linear and nonlinear polarizabilities experience a severe dispersion of the form

\[ \chi^{(n)} \sim (\omega - \omega_2 - i\gamma/2)^{-1} \tag{3} \]

(here \( \gamma \) is the full resonance width); therefore, the function \( W_{\omega \Omega}(k_1) \) defined by (1) and (2) has a rather complex form.

The first component in (1) makes a contribution that is proportional to \( \chi^{(3)''} \) and, for this reason, is noticeable only in close proximity to the resonance \( (\omega_2 \sim \omega_\mu \pm \gamma) \). This component corresponds to ordinary Raman scattering by a "mechanical" (or "Coulomb") phonon of frequency \( \omega_\mu \); it describes a scattering maximum that is independent of the direction of observation. Thus, the frequency-angular spectrum must feature a horizontal straight line ("bridge") (Figure 1.8); this has been corroborated experimentally.

But, on the other hand, the notion of a mechanical phonon is an idealization (suitable only in the region \( k_2 \gg \omega_2/\epsilon \), where the interaction between ion vibrations and the transverse electromagnetic field can be neglected); for this reason, bridge regions do not correspond to real excitations of matter.

The theoretical contradiction here is resolved by taking the second term in (1) into account, as it precisely compensates for the first term in the bridge region (Section 6.6). Experimental bridges are attributed to scattering through 180° and to reflections of the pump and the signal on the crystal faces.
Normal and Anomalous Dispersion

Allowing for the combined action of both terms in (1) helps resolve one other paradox. If we use an ordinary expression for the wave vector in the absorption region, such as

$$\left( \frac{ck_2}{\omega_2} \right)^2 = \epsilon'(\omega_2) \sim \frac{\omega_\mu - \omega_2}{(\omega_\mu - \omega_2)^2 + \gamma^2/4}, \quad (4)$$

which follows from the condition of the minimum denominator of the second term in (1), then the quantity $k_2$ will be limited and the phase-matching condition will be satisfied only in the case of small scattering angles. In such a case, the tuning curve $\theta_1(\omega_1)$, like the function $k_2(\omega_2)$, contains an area with an "anomalous" dispersion in the resonance region (see Figure 1.8).

In experiment, on the other hand, the dispersion usually has the "normal" form, which corresponds to (4) with $\gamma = 0$, and only some of the oscillations give an anomalous dispersion of tuning curves. Both

Figure 1.8. Normal (above) and anomalous types of observed dispersion curves. Scattering with $\theta_1 \sim 0$ and $\omega_2 \sim \omega_0$ (dotted line) is observable due to the reflections on the faces of the crystal.
types of observed resonances are described by (1), with different ratios between the resonance and nonresonance parts of the quadratic polarizability. It turns out that, because of \( \chi^{(2)} \), oscillations that are not active in ordinary Raman scattering with \( \theta_1 \approx 90^\circ \) can appear during scattering at small angles \( (\theta_1 \lesssim 5^\circ) \) in the form of an "anomalous" bending and a local broadening of the tuning curve. These infrared-active oscillations make no contribution to \( \chi^{(3)} \) and, according to (1) and (2), the following scattering coefficient corresponds to them:

\[
W_{\omega \Omega} = \frac{V}{\lambda_1^2} \frac{\chi^{(2)} \chi^{(3)} + 1}{n_2^2 - \varepsilon(\omega_2)}.
\]

This coefficient reaches its maximum in the ordinary case of (4). Equation (5) has a clear physical meaning, viz., thermal fluctuations of the infrared field inside the crystal, which are proportional to (see (4.2.16))

\[
(E^2)_{\omega k} \sim \mathcal{N}_2 \frac{1}{n_2^2 - \varepsilon(\omega_2)},
\]

are transformed into the visible region through the quadratic polarizability \( \chi^{(2)} \) due to the electronic nonlinearity or to the neighboring (in terms of frequency) lattice vibrations. In the latter case, we may draw an analogy with Fermi resonance, where a weak vibration "manifests itself" through its interaction with a strong vibration of a nearby frequency (this term is usually used when the weak vibration is two-photon and its frequency is \( \omega_\mu + \omega'_\mu \)). Equation (6) shows that polariton scattering makes it possible to measure not only the polariton dispersion law (on the basis of the maximum scattering in four-dimensional \( \omega k \) space), but also their lifetime \( \tau \) (or the range of \( \omega \tau \)) associated with \( \varepsilon''(\omega) \) (by the width of the maximum).

It should be noted that the character of the observed dispersion (normal or anomalous) may also depend on the experimental procedure used. If we measure the position of the frequency-dependent scattering intensity maxima \( \omega_1^{\text{max}} \) at different scattering angles \( \theta_1 \), then the function \( \omega_1^{\text{max}}(\theta_1) \)—even for \( \chi^{(3)} = 0 \), in accordance with (6)—will have the "normal" form of two repulsive hyperbolas.
The Linearization Effect

The phenomenological formulas (1) and (2) describe one other effect. Even in the early experiments, in which parametric scattering was observed in lithium niobate crystals, it was noted that the scattering vanished as the idler frequency approached the infrared absorption region ($\nu_2 \sim 1400 \text{ cm}^{-1}$). This phenomenon was naturally attributed to the negative influence of the idler wave's damping on the efficiency of parametric interaction. However, according to the fluctuation-dissipation theorem (Section 2.4), fluctuations increase with an increase in absorption; thus, only the tuning curve $\omega_1(\theta_1)$ should be broadened, the integral intensity (in terms of $\omega_1$ or $\theta_1$) remaining unchanged. This phenomenon is described by (6). Indeed, it was established in subsequent experiments that the scattering returned after a "dip" at the frequency of $1400 \text{ cm}^{-1}$ even though $\alpha_2$ continued to grow. It has now been established beyond any reasonable doubt that the "dip" is caused by interference between the electronic $\chi^{(2)}_{\infty}$ and the electronic-vibrational $\chi^{(2)}_{\text{res}}$ nonlinearities, which results in the "linearization" of the crystal's susceptibilities ($\chi^{(2)}_{\infty} + \chi^{(2)}_{\text{res}} = 0$) at certain frequencies. The same "dips" are observed on tuning curves near many lattice resonances $\omega_\mu$. This effect is analogous to the zero passage of the linear dielectric permeability at longitudinal resonance frequencies.

1.3 Four-Photon Scattering and Nonlinear Spectroscopy

As the majority of materials, such as gases, liquids, glasses, and centrosymmetrical crystals, do not have a well-defined polar direction, their macroscopic quadratic polarizability is zero and three-photon effects, including parametric scattering and scattering by polaritons, are impossible or negligible (because of magnetic interactions). However, in the next order of perturbation theory with respect to the pump amplitude, four-photon effects do appear and these are phenomenologically described by the cubic polarizability $\chi^{(3)}$. This quantity links four polar vectors and is a fourth rank tensor on whose
existence the symmetry of the medium does not impose any general exclusions.

As in the case of parametric and polariton scattering, four-photon elementary processes are conveniently classified on the basis of transparency criteria (with respect to one-photon processes, i.e., in the first order by the pump). The cubic polarizability is a function of three independent frequency arguments; on the basis of relations between these frequencies and the natural (Bohr) frequencies of the molecules, it is possible to identify a good many processes or, to put it another way, transitions. Figure 1.9 gives a schematic of various multiphoton transitions. It will be seen that all transitions can be divided into two classes—some change the state of both the molecule and the field; others (known as parametric or coherent) alter only the state of the field. In terms of polarizabilities, the imaginary parts $\chi^{(2n-1)}$ correspond to nonparametric $n$-photon transitions.

Hyperparametric Scattering$^{9-24}$

In this case, by definition, matter is transparent at all the frequencies involved in the scattering process. Hyperparametric scattering (HPS) or light-by-light scattering (the latter term can naturally be applied to three-photon parametric scattering also) may be interpreted as the result of the decay of two pump photons into a photon pair with other frequencies, directions, and polarizations (Figure 1.9). The following conservation laws must be obeyed in this instance:

$$\omega_1 + \omega_2 = \omega_3 + \omega_4, \quad k_1 + k_2 = k_3 + k_4.$$ \hspace{1cm} (1)

As a rule, degenerate pumping is used ($\omega_3 = \omega_4$ and $k_3 = k_4$) and scattering is also observed in the anti-Stokes region. Condition (1), together with the variance of the refractive index, determines the frequency-angular spectrum of the scattered light. If the linear dispersion is neglected, then (1) determines the phase-matching surface in the form of an ellipsoid with an interfocal distance $|k_3 + k_4|$. When $k_3 \parallel k_4$, the light is scattered only in the forward direction, while, when $k_3 = -k_4$, the scattering is isotropic and elastic.

It is easy to estimate the intensity of hyperparametric scattering using the relations obtained in Section 1.1 if we make the following
INTRODUCTION

Figure 1.0. Elementary multiphoton processes. The thin arrows correspond to spontaneous radiation. The processes a, g, i, j, and k are parametric, leading to a quantum correlation between different frequency modes and photon superbunching; the same effect is generated by two-photon radiation in d, e, and f; a and g are parametric and hyperparametric scatterings; b, c, and i are Stokes, anti-Stokes, and coherent Raman scattering; e is the cascade process; f is the interference of one- and two-photon transitions, leading to the appearance of the third moment of the field; h is hyper-Raman scattering; j and k are one- and two-photon resonance fluorescence.

In the first nonvanishing approximation, the HPS intensity is proportional to the square of the pump intensity \([S_{1} \sim S_{2} S_{4}]\); thus, one must use pulsed and/or focussed pumping to observe it. If a very powerful pump is used, four-photon parametric superluminescence is observed, most of which is in the forward direction and which spans the entire spectrum up to the double pump frequency almost without a break.
Cascaded Hyperparametric Scattering\textsuperscript{19–24}

In noncentrosymmetric crystals, a scattering that is quadratic in terms of pump intensity may also occur because of the two-stage (cascade) processes in which the light of ordinary parametric scattering interacts with the pump once again before leaving the crystal:

\[ \omega_3 \rightarrow \omega_2 + \omega'_1, \quad \omega_3 + \omega'_1 \rightarrow \omega_1. \]  

This process is readily observable in the case of double phase-matching, when

\[ k_3 = k_2 + k'_1, \quad k_3 + k'_1 = k_1, \]  

which obtains in exceptional cases only. However, in principle, cascade HPS may have a significant value both at the non-phase-matched first stage and the phase-matched second stage (or vice versa). Such processes lead to the imposition of a fundamental limitation on the sensitivity of three-photon frequency converters and quantum counters. The role of the second pump can be played by an acoustic wave.\textsuperscript{25}

Hyper-Raman Scattering\textsuperscript{26}

The cubic susceptibility of the medium has the order $\chi^{(3)} \sim \gamma M_0$, where $\gamma$ is the cubic hyperpolarizability of a single molecule and $M_0$ is the molecular concentration. When the natural frequencies of the material $\omega_\mu$ (collective or individual) coincide with the frequencies of the photons, the material's nonlinearity goes up sharply.

Thus, with $\omega_2 \rightarrow \omega_\mu$, where $\omega_\mu$ is the frequency of molecular vibrations, hyperparametric scattering continuously develops into hyper-Raman scattering (HRS; Figures 1.9h and 1.10). This effect, which is sometimes also called three-photon Raman scattering, is attributed to the decay of two pump photons into a Stokes photon with a frequency $\omega_4 = 2\omega_3 - \omega_\mu$ and a phonon with a frequency $\omega_\mu$. If nuclear vibrations within the molecule involve a change in the dipole moment, then hyper-Raman scattering by polaritons proportional to the imaginary
part (cf. (1.2.1)) of the quantity

\[
\Delta \epsilon_1^{(4)} = 4\pi |E_3|^4 \left[ \chi^{(5)} + \frac{4\pi \chi^{(3)}_2}{\varepsilon'(\omega_2)} \right],
\]

(5)

where \(\chi^{(5)}\) is a fifth-order polarizability with respect to the field, should be observed.

Biphonon Radiation in Raman Scattering

If \(\omega_2 \to \omega_3 \pm \omega_\mu\), then hyperparametric scattering continuously turns into Raman scattering (Figures 1.9b, c, i, and 1.10). The Raman resonance of cubic polarizability describes, apart from Raman scattering, a number of other interesting phenomena—such as inverted Raman scattering, which features not scattering but a corresponding stimulated amplification effect of the Stokes wave or absorption of the anti-Stokes wave. Another example is the resonance frequency conversion \(2\omega_3 - \omega_2 \to \omega_1\) (stimulated coherent anti-Stokes Raman scattering—CARS) which forms the basis of what is known as active spectroscopy.\(^{27}\)

These stimulated effects can be described by means of a semiclassical theory of radiation. Quantum optics predicts a new, but as yet undiscovered, feature of Raman scattering at small angles in which the phase-matching condition (1) is satisfied.\(^{28,29}\) In Section 7.3, it is shown that, with \(\hbar \omega_\mu \gg \kappa T\), anti-Stokes photons are only radiated si-

---

**Figure 1.10.** A rough representation of the frequency-angular spectrum of light being scattered by matter. (1) Stokes Raman scattering. (2) Rayleigh scattering. (3) Coherent Raman scattering (or resonance hyperparametric scattering or spontaneous CARS). (4) Hyperparametric scattering. (5) Hyper-Raman scattering. (6) Hyperpolariton scattering. (7) Hyper-Rayleigh scattering.
multaneously with the Stokes photons. This effect may be interpreted as resonance hyperparametric scattering (Figures 1.9i and 1.10).

Resonance Fluorescence

One other type of one-fold resonance of cubic polarizability is possible: one with $\omega_3 \sim \omega_\mu$, where $\hbar \omega_\mu$ is the excitation energy of an electron in an atom or molecule. In this case, linear susceptibility and Rayleigh scattering increase simultaneously, resulting in resonance fluorescence.\(^{30,31}\) Resonance fluorescence of single atoms has made it possible to observe photon antibunching.\(^{32}\)

By allowing for the resonance cubic polarizability, one can describe the effects of saturation and radiation of photon pairs with the following frequencies:

$$\omega_2 = \omega_\mu \equiv \omega_3 - \delta, \quad \omega_1 = 2\omega_3 - \omega_\mu = \omega_3 + \delta. \quad (6)$$

However, as in the case of polariton scattering, idler photons are quickly reabsorbed; thus, biphotons can only be radiated by the surface layers of the sample. In the next orders of perturbation theory, radiation arises at the frequencies $\omega_3 \pm 2\delta$, etc. Analogous effects of the generation of higher components are observed in stimulated Raman scattering. The radiation of frequency $\omega_1$ in (6) can be regarded as the result of hyper-Raman scattering by electronic excitation.

It will be noted that the ordinary saturation effect, which also is described to a first approximation by cubic polarizability, is used in a variant of Doppler-free spectroscopy known as saturation spectroscopy.\(^{33}\)

Two-Photon Resonance Luminescence

Finally, the fourth type of cubic polarizability resonance occurs with $2\omega_3 \sim \omega_\mu$. In addition to two-photon pump absorption, resonance hyperparametric scattering and biphoton radiation should also be observed (Figure 1.9k). The use of a standing stationary pump wave ($k_3 = -k_4$) in the case of two-photon absorption in gases dispenses with the Doppler broadening of spectral lines.\(^{33}\) A similar line-narrowing effect should also be observed in two-photon resonance lumi-
nescence in the case of nearby frequencies $\omega_1$ and $\omega_2$. The sensitivity of this variant of Doppler-free spectroscopy can be enhanced by means of a coincidence detector.

**Nonlinear Spectroscopy**

It is clear from the foregoing discussion that the measurement of the three-dimensional frequency dispersion of cubic polarizability $\chi^{(3)}(\omega_2, \omega_3, \omega_4)$ provides extensive spectroscopic information. The choice of the type of resonance and the type of effect, whether spontaneous or stimulated, makes it possible to optimize the resolution and sensitivity of the method employed. Interesting examples of stimulated single- and double-resonance cubic effects were observed in vapors of alkali-halogen metals. As in the case of linear spectroscopy, it is possible to use the reactive (phase) effects described by the real part $\chi^{(3)}$, as well as to change the polarization of the output fields at a resonance.

**1.4 The Basic Concepts of Statistical Optics**

Thus far we have dealt only with the intensity (spectral brightness) of radiation and with its dependence on the frequency and direction of observation. Although an important characteristic of radiation, intensity does not provide complete statistical information on the electromagnetic field.

In this section, we shall examine a series of typical experimental schemes in order to analyze actually observable parameters and the corresponding theoretical values of the correlation functions $G_i^{(n)}$ (the index $i$ replaces the set of arguments $r_i, t_i, \alpha_i$).

Since the vast majority of observable optical effects do not require field quantization for an explanation, we shall confine ourselves to the simpler and more graphic classical theory citing, where necessary, the results of the quantum theory (Chapter 3).
The Analytic Signal

It is convenient, in statistical optics, to replace the real field strength $E_\alpha(x,t)$ with the complex function $E_\alpha^{(+)}(x,t)$, which is known as the analytic signal or the positive-frequency part of the field. This function is unequivocally defined by the following equality (the arguments $\alpha$ and $x$ are omitted):

$$E^{(+)}(t) \equiv \int_0^\infty d\omega e^{-i\omega t} E(\omega),$$

where the Fourier transform of the real field is

$$E(\omega) \equiv \frac{1}{2\pi} \int_{-\infty}^\infty dt e^{i\omega t} E(t) = E^*(-\omega).$$

By definition, the spectrum of the analytic signal $E^{(+)}(t)$ contains only positive frequencies $\omega > 0$, while the spectrum of the complex-conjugate (negative-frequency) function $E^{(-)}(t) = E^{(+)}(t)^*$ contains only negative frequencies. The relation

$$E(t) = E^{(+)}(t) + E^{(-)}(t) = 2 \text{Re} E^{(+)}(t)$$

follows from this definition (Figure 1.11). In the case of a quasi-monochromatic ("modulated") field with a narrow spectrum $\Delta \omega$, it is convenient to introduce a "slow" complex amplitude $E_0(t)$ that is defined by the equation

$$E(t) \equiv \frac{1}{2} E_0(t) e^{-i\omega t} + \text{c.c.},$$

where $\omega$ is a certain mean frequency. The modulus $|E_0(t)|$ is called the envelope and the argument is called the slowly changing phase. The spectrum of $E_0$ occupies the interval $\pm \Delta \omega/2$ in the zero-frequency region; given $\Delta \omega \ll \omega$, we may obviously assume

$$E^{(+)}(t) \approx \frac{1}{2} E_0(t) e^{-i\omega t}. $$
The readings of optical detectors—photomultipliers, bolometers, photographic films, etc.—depend on the squares (or even higher degrees) of the field averaged for a certain time interval $T$ over the time lag of the detector’s response (for the moment, we abstract ourselves from the finiteness of its spatial dimensions). Let $\Delta \omega \ll 1/T \ll \bar{\omega}$ (i.e., the envelope changes little during the detector’s response time, encompassing as it does many periods of the optical field); the detector’s readings will then be proportional to the “instantaneous intensity” (we omit here the proportionality factor, which—in a plane wave—equals $c/2\pi$):

$$I(t) \equiv \frac{1}{2} \overline{E^2(t)} \approx |E^{(+)}(t)|^2 = \frac{1}{4} |E_0(t)|^2$$

(6)

The use of the analytic signal here automatically dispenses with those components which oscillate with a double optical frequency.

Random Intensity

The instantaneous intensity $I(t)$ at the point $r$ changes in a random field, fluctuating both in time and in space. (Generally speaking, we should examine the random tensor $E_\alpha^{(-)} E_\beta^{(+)}$, where $\alpha = x, y, z$, or, in the case of directional radiation, $\alpha = x, y$.) Often only the most
elementary field characteristics are measured—the mean intensity at a given point
\[ \langle I \rangle = \left\langle E^{(-)}E^{(+)} \right\rangle \] (7)
and the average square of the intensity
\[ \langle I^2 \rangle = \left\langle (E^{(-)})^2E^{(+)} \right\rangle \] (8)
or the variance
\[ \Delta I^2 \equiv \langle I^2 \rangle - \langle I \rangle^2. \] (9)

The parameters (7–9) can, in principle, be measured by determining the mean value and variance of the readings of a wide-band single-quantum photodetector (see below). The readings of an n-quantum detector immediately gives the nth moment in \( \langle I^n \rangle \equiv G^{(n)} \).

Let us assume that the field is stationary (so that the mean values in (7–9) are time independent) and ergodic. The angle brackets indicate averaging over time or over an ensemble (by means of the distribution function \( P(I) \)).

Turning to quantum theory, the quantity \( E^{(+)} \) is replaced by the operator \( \hat{E}^{(+)} \), which is expressed through the photon annihilation operators \( a_k \), and \( E^{(-)} \) is replaced by the operator \( \hat{E}^{(-)} \), which is expressed through the creation operators \( a_k^\dagger \). In the process, the sign of complex conjugation is replaced by the sign of Hermitian conjugation \( \ast \), while the angle brackets indicate quantum averaging by means of the wavefunction or the density matrix. Most of the quantum states of the field also allow averaging by means of \( P(z) \), the quasiprobability function, which is analogous to classical averaging. In switching to quantum averaging, the order of writing operators becomes important; the sequence adopted in (7–8) is called normal. Within the framework of classical theory, one may, of course, replace the normal moment \( \left\langle E^{(-)}E^{(+)} \right\rangle \) by \( \left\langle \left| E^{(+)} \right|^2 \right\rangle \).

In the vast majority of cases, the optical field is excited by a multiplicity of independent sources with random amplitudes and phases; for instance, in the case of the thermal radiation of a heated material or a quantum mechanical amplifier or in the case of a multimode laser with independent modes. In such cases, the distribution of the
complex amplitude $E_0 \equiv E_0^e + iE_0^m$ is normal (Gaussian) with independent $E_0^e$ and $E_0^m$, while the intensity distribution is exponential (Figure 1.12):

$$P(I) = \langle I \rangle^{-1} \exp(-I/\langle I \rangle).$$

(10)

Thus, the mean intensity $\langle I \rangle$ completely determines the statistics of a stationary chaotic field (at one point and for one type of polarization). It should be noted that, more often than not, zero intensity occurs: $P(0) \geq P(I)$. The moments and variance of intensity can easily be found by means of (10):

$$G_T^{(n)} = n! \langle I \rangle^n, \quad \Delta I_T^2 = \langle I \rangle^2$$

(11)

(the index $T$ corresponds to the chaotic field).

Another characteristic case is the emission of a single-mode laser with a stabilized amplitude. In this instance,

$$P(I) = \delta(I - I_0), \quad G^{(n)} = I_0^n, \quad \Delta I = 0.$$  

(12)

Figure 1.12. Two basic types of the state of the field. An approximate representation of the change of the field in time from a classical point of view (a) and the corresponding distribution functions—both classical (b) and quantum (c).

Top: The quasimonochromatic field of thermal radiation or a quantum amplifier or generator (below threshold) when both amplitude and phase fluctuate.
Bottom: The field of a single-mode laser in which amplitude fluctuations are suppressed by the saturation effect.
Equations (10—12) do not take the discrete character of possible values of the field’s energy—i.e., its photon structure—into account; for this reason, they are suitable only for those classical fields which have a degeneracy factor (see below) considerably greater than unity. We shall examine the general case in Chapter 3. For the present, we shall note that, in the quantum theory, the continuous distribution (10) is replaced by a discrete exponential distribution, while the continuous distribution in (12) is replaced by Poisson’s distribution (Figure 1.12).

Correlation Functions

The distributions and intensity moments considered above give us no information on the correlation between the fields in adjacent points of space-time or that between the various Cartesian components of the field. Complete information is provided by a series of multidimensional distributions or tensor correlation functions (CF). The latter, according to Glauber,\(^1\) are defined in the following fashion (using a notation that is also suitable for the quantum consideration):

$$G^{(n)}_{1\ldots 2n} \equiv \langle E_1(-) \cdots E_n(-) E_{n+1}(+) \cdots E_{2n}(+) \rangle. \quad (13)$$

Here the lower indices replace the totality of arguments, for instance $E_1 \equiv E_{\alpha_1}(r_1,t_1)$. For simplicity’s sake, we consider only those correlation functions with even numbers of fields since, in optics, moments of the $\langle E_1 E_2 E_3 \rangle$ type are usually equal to zero and, besides, they are not easy to detect (see, however, Section 5.3).

The correlation functions (CFs) of the stationary field are invariant with respect to the reference point, i.e., relative to the substitution of temporal arguments

$$t_1, \ldots, t_{2n} \rightarrow t_1 + \Delta t, \ldots, t_{2n} + \Delta t, \quad (14)$$

where $\Delta t$ is arbitrary (it is convenient to set $\Delta t \equiv -t_1$).

As a result, $G^{(n)}$ is dependent on $2n-1$ temporal arguments while its Fourier image—the spectral CF $\tilde{G}^{(n)}$—is dependent on $2n-1$ frequencies. For instance, the first order CF (CF-1) has the form $G(r_1, r_2, \tau)$; it is called the function of mutual coherence of the field.
at points $r_1$ and $r_2$, while its Fourier image $\tilde{G}(r_1, r_2, \omega)$ is known as the mutual spectral density. At $r_1 = r_2$, CF-1 $G(r, \tau)$ is called the autocorrelation function, while $\tilde{G}(r, \omega)$ is called the spectral density. The traditional polarization characteristics of the directional field, such as polarization degree or Stokes parameters, are also expressed through CF-1, the tensor indices being taken into account (for details, see Reference 8). When all the arguments are coincident, the CFs cross into intensity moments (we omit the tensor indices),

$$G_{1\ldots1}^{(n)} = \langle |E^+(r_1, t_1)|^{2n} \rangle = \langle I^n(r_1) \rangle. \quad (15)$$

Among all the possible statistical field models, the Gaussian model, in which all the CFs are expressed in terms of CF-1, holds a special position:

$$G_{1\ldots n1'\ldots n'}^{(n)} = \sum' G_{11'}^{(1)} \cdots G_{nn'}^{(1)}, \quad (16)$$

where $\sum'$ stands for the sum of all $n!$ permutations of the primed indices. For instance,

$$G_{1234}^{(2)} = G_{13}^{(1)} G_{24}^{(1)} + G_{14}^{(1)} G_{23}^{(1)}. \quad (17)$$

Complete information on the Gaussian (chaotic, thermal) field is provided by the first-order correlation function $G_{\alpha\beta}(r_1 r_2, \tau)$ or by the mutual spectral density $\tilde{G}_{\alpha\beta}(r_1, r_2, \omega)$.

For a rough description of the quasimonochromatic directional nonpolarized Gaussian field, it is sufficient to determine the intensity $G_\nu(r)$ and the scale of coherence $\tau_{\text{coh}} \sim 1/\Delta \omega$, $\rho_{\text{coh}}$ for both polarization types ($\nu = 1, 2$) at all points. The meaning of these parameters will be explained below by means of simple measuring procedures.

**Temporal Coherence**

Let us examine the field $E'(t)$ at the output of Michelson's interferometer (Figure 1.13). It consists of two components differing by a certain delay time $\tau = t' - t$:

$$E'(t) = \frac{1}{2} [E(t) + E(t')],$$
where $E(t)$ is the field in a plane wave at the input of the interferometer (without allowing for the delay common to both beams). In accordance with (6), we find the intensity at the output:

$$I'(t) = \frac{1}{4} \left[ I(t) + I(t') + 2 \text{Re} E^(-)(t)E^+(t') \right].$$

It follows from this that, in the case of stationary radiation,

$$\langle I' \rangle = \frac{1}{2} \left[ \langle I \rangle + \text{Re} G(t) \right] = \frac{1}{2} \langle I \rangle \left[ 1 + \text{Re} g(t) \right], \quad (18)$$

where

$$G(\tau) \equiv \langle E^-(0)E^+(\tau) \rangle = \langle E^0_0(0)E^0_0(\tau) \rangle e^{-i\omega \tau} \quad (19)$$

is the autocorrelation function for the field at the output and is linked, via the Wiener–Hinchin theorem, with the spectral density $\tilde{G}(\omega)$ through the Fourier transform; $g(\tau) \equiv G(\tau)/\langle I \rangle$ is the normalized CF; and $\langle I \rangle = G(0)$. Note that, in optics, CFs are usually normalized by mean values and not by standard deviations $\Delta I$.

According to (18), the dependence of intensity at the output of the interferometer on the delay time determines the real part of CF-1. It can be shown that the function $G(\tau)$ (as well as $E^+(\tau)$) is analytic in the lower half-plane $\text{Im} \tau < 0$; therefore, its real and imaginary parts are linked through the Hilbert transformation and, in principle, the spectrum can be restored on the basis of the interference picture. This method underlies Fourier spectroscopy. On the other hand, the

![Figure 1.13. A schematic of Michelson’s interferometer, which is used to measure time coherence.](image)

Figure 1.13. A schematic of Michelson’s interferometer, which is used to measure time coherence.
interference of the field under study with the reference coherent wave serves as a source of information in holographic interferometry.

Figure 1.14 shows an approximate interference picture for the case of a single spectral line. The relative swing of oscillations exhibited by the output intensity is called the visibility of the interference picture or, alternatively, the degree of coherence. According to (18) and (19), the visibility practically coincides with $|g(\tau)|$.

If we define the coherence time by the condition $|g(\tau_{\text{coh}})| = 1/2$, then it follows from the properties of the Fourier transform that $\tau_{\text{coh}} \sim 2\pi/\Delta \omega$. Hence, the length of longitudinal coherence $r_{\text{coh}}$ has the order of the inverse width of the spectrum in cm$^{-1}$:

$$l_{\text{coh}} \sim \frac{2\pi c}{\Delta \omega} = \frac{1}{\Delta \nu}.$$  \hspace{1cm} (20)

For instance, for lines with a natural broadening in the visible range, $\tau_{\text{coh}} \sim 10^{-8}$ sec and $l_{\text{coh}} \sim 50$ cm. Descriptively, this is the mean length of the wave train emitted by the atom during spontaneous emission. In the case of a single-mode laser estimate based on Townes’ formula (4.5.30), the coherence length is on the order of a light second.

Figure 1.14. A rough representation of the dependence of the mean intensity at the output of Michelson’s interferometer on the position of the mobile mirror ($g$ is the normalized autocorrelation function at the input).
Spatial Coherence

The correlation of fields at two different points, \( r_1 \) and \( r_2 \), separated by a space-like interval can be measured using the Young interferometer—a screen with two holes that is set up perpendicular to the direction of the emission from the source (Figure 1.15).

The interference picture at an arbitrary point behind the screen is defined, by analogy with (18), by a CF-1 of the general form

\[
G_{12}(\tau) \equiv G(r_1, r_2, \tau) = \langle E_1^(-)(t)E_2^+(t) \rangle ;
\]

on the axis of symmetry, it is defined by the function \( G_{12}(0) \). The coherence radius \( \rho_{coh} \) is determined by the distance between the holes at which visibility at the center drops to 50 percent.

An ideal laser operating on a single transverse mode emits plane or spherical waves for which \( \rho_{coh} = \infty \). On the other hand, for the case of a multimode laser—or, in general, of a chaotic (thermal) source—\( \rho_{coh} \) is determined at large distances \( (z \gg a^2/\lambda) \) by the transverse dimension of the source \( a \) and by the distance \( z \) to it (Figure 1.15):

\[
\rho_{coh} \sim \frac{\lambda z}{a} = \theta_d z = \frac{\lambda}{\theta_a} ;
\]

where \( \theta_d \) is the angle of diffraction and \( \theta_a \) is the angular size of the source. The radius of transverse coherence due to diffraction increases with propagation. The form of the phase front evens out in the process, approaching the spherical. The Van Cittert-Zernike theorem,\(^2,\(^8\)

![Figure 1.15. A schematic of Young's interferometer, which is used to measure the radius of transverse coherence.](image-url)
which describes this phenomenon quantitatively, states that the dependence of \( CF-1 \) on \( (r_1 - r_2)_\perp \) is the Fourier image of the brightness distribution across the cross-section of the source.

This relationship (22) enabled Michelson to determine the angular diameters of certain stars for which \( \theta_a \geq 10^{-7} \) rad and \( \rho_{\text{coh}} \leq 10 \) m by means of his "stellar" interferometer. At smaller \( \theta_a \), the coherence radius is determined, in contrast, by the distortions of the wave front induced by the atmosphere. The Brown-Twiss intensity interferometer is free of this defect.

Coherence Volume and the Degeneracy Factor

The volume of coherence is the product of coherence area by coherence length, \( \rho_{\text{coh}}^2 l_{\text{coh}} \). For the far field of a chaotic source, it follows from (22) that

\[
V_{\text{coh}} = \frac{\lambda^4}{\Delta\lambda\Delta\Omega_a},
\]

where \( \Delta\lambda = \lambda^2/l_{\text{coh}} \) and \( \Delta\Omega_a \equiv (a/z)^2 \) is the solid angle within which the source is visible.

An important dimensionless statistical parameter of radiation is the degeneracy factor—the mean field energy (of a single polarization) in units of \( \hbar\bar{\omega} \), which is contained in the coherence volume, i.e., the number of photons \( \langle N \rangle \) in the volume \( V_{\text{coh}} \) or, to put it another way, the number of photons intersecting the coherence area in coherence time:

\[
\delta \equiv \langle E \rangle_{\text{coh}} / \hbar\bar{\omega}.
\]

Let us express \( \langle E \rangle_{\text{coh}} \) and \( \delta \) through the spectral brightness of radiation (of a definite polarization):

\[
\langle E \rangle_{\text{coh}} = S_{\omega\Omega} \Delta\omega \Delta\Omega_{\text{coh}} \rho_{\text{coh}}^2 = 2\pi\lambda^2 S_{\omega\Omega},
\]

\[
\delta = \frac{\lambda^3}{\hbar c} S_{\omega\Omega} \equiv \langle N \rangle.
\]

Thus, the degeneracy factor is equal to spectral brightness in units of \( \hbar c/\lambda^3 \). In other words, the mean number of photons in a coherence
§1.4 THE BASIC CONCEPTS OF STATISTICAL OPTICS

volume is the same as the mean number of photons in one mode \( \langle N \rangle \). In an equilibrium field, \( \delta = \mathcal{N}_T = \frac{1}{\exp(\hbar \omega / \kappa T) - 1} \) (in a nonequilibrium field, \( T \) should be taken to mean the brightness temperature \( T_{\text{eff}} \)).

The degeneracy factor \( \delta \), besides being a convenient dimensionless measure of spectral brightness, also determines the applicability of classical statistics—at \( \delta \lesssim 1 \) (i.e., \( \kappa T \lesssim \hbar \omega \)), the photon structure of the field becomes significant.

Let us split the space in the far field of the chaotic quasimonochromatic source into cells with a volume of \( V_{\text{coh}} \). Radiation in any one pair of points belonging to a cell is, by definition, mutually coherent and we may regard the field within the cell as approximately single-mode, i.e., assume it to be a spherical monochromatic wave with a particular amplitude \( |E_0| \) and phase \( \phi \). During passage from one cell to another, \( |E_0| \) and \( \phi \) will fluctuate in a random fashion. Thus, the spatial distribution of the chaotic field forms an ensemble of states of the harmonic oscillator.

Let fields belonging to different coherence volumes be noncorrelated, or even independent. Consequently, by virtue of the central limiting theorem, the energy distribution in the volume \( V \), which is much greater than \( V_{\text{coh}} \), will be Gaussian with a variance inversely proportional to the number of cells.

However, in the quantum region, when \( \delta \lesssim 1 \), the Gaussian distribution for the multimode field is replaced by a Poisson distribution (Section 3.4).

Table 1.1 presents some of the characteristic types of distributions of field energy or the number of photons. The states of the field, which are called, for convenience, \( K \)-photon light, have no classical analogs and exhibit photon antibunching or superbunching effects (Section 3.4). \( K \)-photon light consists, roughly speaking, of groups of \( K \) simultaneously emitted photons (see Figure 3.5d for 2-photon light).

### Photocount Statistics. Mandel's Formula

The law governing the distribution of intensity in a field at a single "point" can be measured by means of a photomultiplier operating as a photon counter. The mean light intensity \( \langle I \rangle \) is set low enough to
Table 1.1
The Main Types of Distributions of the Photon Number and Field Energy

<table>
<thead>
<tr>
<th>State modes</th>
<th>Quantum Theory</th>
<th>Classical Theory</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coherent</td>
<td>Poisson</td>
<td>Poisson</td>
</tr>
<tr>
<td>Chaotic</td>
<td>Bose-Einstein (geometric)</td>
<td>Poisson</td>
</tr>
<tr>
<td>K-photon (mixed)</td>
<td>( P_0 = 1 - \langle N \rangle / K )</td>
<td>Poisson</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Number of independent modes</th>
<th>1</th>
<th>( \gg 1 )</th>
<th>1</th>
<th>( \gg 1 )</th>
</tr>
</thead>
</table>

4.6 INTRODUCTION

§1.4

Table 1.1
The Main Types of Distributions of the Photon Number and Field Energy

ensure that individual nonoverlapping current pulses are generated at the output of the photomultiplier (Figure 1.16). Repeated counts of the number of pulses appearing within a specified interval \( T \) makes it possible to establish the law governing the distribution \( P(m) \) of the number of initial photoelectrons knocked out by the incident light from the photocathode (the total measurement time should, naturally, greatly exceed the coherence time \( \tau_{coh} \)).

Let us find the relationship between the statistics of the photo-counts and those of the field. Let \( T \ll \tau_{coh} \) and \( A \ll A_{coh} \); we may then neglect the change in field intensity occurring per sampling session \( T \) and the surface of cathode \( A \). In this instance, the distribution \( P(m) \) will be determined through \( P(I) \) irrespective of the detection volume \( V_{det} \equiv cTA \). Here \( \tau_{coh} \) and \( A_{coh} \) are the characteristic scales of field fluctuations and the inequalities allow us to regard the detector as being "point" or "single-mode," i.e., measuring one degree of field freedom (for this the detector should also measure a single type of polarization).

Let us first assume that the field intensity \( I \) is constant, i.e., that it does not vary from sampling session to sampling session. Significantly, the number of photoelectrons in a sampling is random and unpredictable even in this case, in accordance with the principles of quantum mechanics. The process of measuring the field energy inevitable introduces an additional Poisson stochasticity into the de-
§1.4  THE BASIC CONCEPTS OF STATISTICAL OPTICS  47

Figure 1.16. The relationship of field statistics and photoelectron statistics from a classical point of view ($E$ is the field amplitude and $i$ is the current of the photomultiplier tube).

(a) In the case of a field with a constant amplitude, the number of photoelectrons $m$ arising in a certain time interval $T$ fluctuates according to a Poisson distribution.

(b) Field fluctuations when $T < \tau_{coh}$ cause additional fluctuations of electrons (bunching).

(c) A multimode ($T > \tau_{coh}$) detector averages out field fluctuations and no bunching effect is observed.

tector's readings (if we exclude the unrealistic case of a 100 percent quantum efficiency of the photomultiplier and a pure energy state of the field with a definite number of photons). Whether we use the semiclassical or wholly quantum (Section 3.5) perturbation theory, we can only determine the probability $W_1 \Delta t \sim I$ of the ionization of
one cathode atom within a short interval $\Delta t$.

In the case of a sufficiently low $\Delta t$, the probability of the ionization of any one of $N$ independent atoms will be $N$ times greater and also proportional to $I$:

$$W = NW_1 \equiv \frac{\alpha}{I}. \quad (27)$$

Here $\alpha$ is the proportionality constant, which can be presented in the form

$$\alpha = \eta V_{\text{det}}/2\pi \hbar \omega, \quad (28)$$

where $V_{\text{det}} \equiv cTA$ is the effective detection volume, $\eta \equiv \sigma l N_0 = \sigma N/A$ is the quantum yield of the thin cathode of $l$ thickness, $\sigma$ is the ionization cross-section, and $N_0 = N/Al$ is the concentration of atoms (we consider $\sigma$ to be constant within the limits of the spectral width of the field).

We assume that all time moments within the limits of $T$ are equivalent (i.e., that a "pure sinusoid" is incident on the cathode) and that an electron may appear with equal probability $\alpha I \Delta t/T$ during any short interval $\Delta t$. This stochastic model, as can easily be shown (see, e.g., Reference 2), leads to a Poisson distribution with the parameter $\alpha l$:

$$P(m | I) = C \frac{(\alpha l)^m}{m!}, \quad C \equiv e^{-\alpha I}. \quad (29)$$

To allow for the intensity fluctuations from sampling session to sampling session, one must average (29) by means of the distribution $P(I)$:

$$P(m) = \int_0^\infty dIP(m | I)P(I) \equiv \langle P(m | I) \rangle. \quad (30)$$

As a result, we arrive at Mandel's semiclassical formula for the distribution of photocounts, i.e., for the probability of detection of $m$ pulses at the output of the photomultiplier:

$$P(m) = \frac{1}{m!} \langle (\alpha l)^m e^{-\alpha I} \rangle. \quad (31)$$
The quantum theory, which was chiefly developed by Glauber,\(^1\) gives an expression in the same form, the only difference being that the probability \(P(I)\) is replaced by a quasiprobability, a function that, in certain states of the field, has negative values and takes on singular forms (of the type of derivatives of the \(\delta\) function).

The expansion of (31) into a series makes it possible to express \(P(m)\) in terms of the moments of intensity \(G^{(k)} \equiv \langle I^k \rangle\) with \(k \geq m:\)

\[
P(m) = \sum_{n=0}^{\infty} (-1)^n \frac{\alpha^{m+n}}{m!n!} G^{(m+n)}.
\]

With the detection volume sufficiently small \((\alpha \sim TA \to 0)\), one may, as a rule, allow for only the first member of this series (i.e., neglect the exponent in (31). In this case, \(P(m)\) can be represented in the form

\[
P(m) = \frac{T^m}{m!} W_1^m = \frac{(NT)^m}{m!} \langle W_1^m \rangle,
\]

where \(W_1^m\) is the \(m\)th derivative of \(P(m)\) over \(T\), \(W_1\) is the ionization probability of one atom in unit time, and \(N\) is the total number of atoms in the detector.

Mandel's formula (31) describes an ensemble of samplings distinguished by time shifts (thanks to the assumed stationarity and ergodicity of the field). It will be seen that, in the case of homogeneous (and "spatially ergodic" along the \(x\) and \(y\) axes) radiation propagating approximately along the \(z\) axis, the same formula describes an ensemble of samplings distinguished by a shift in the \(x, y\) plane. In principle, this enables us to investigate the statistics of nonstationary fields "in real time" by means of a great number of counters distributed at various points in the cross-section of the light beam over an area far greater than \(A_{\text{coh}}\).

Furthermore, it is clear from the derivation of (31) that it also holds in the case of an arbitrary detector volume \(V_{\text{det}} = cTA\), if by \(I\) we mean an intensity averaged over \(V_{\text{det}}:\)

\[
\bar{I}(r,t) = \frac{1}{AT} \int dxdy'dt' I(x',y',z,t),
\]

where the limits of integration, \(x \pm a/2, y \pm b/2,\) and \(t \pm T/2,\) are
determined by detector dimensions. In this case, the distribution \( P(I) \) should be replaced by \( P(\bar{I}) \). In the extreme case, where \( A \gg A_{\text{coh}} \) and/or \( T \gg \tau_{\text{coh}} \) ("multimode" detection), the fluctuations of \( I \) are completely smoothed out: \( P(\bar{I}) = \delta(\bar{I} - \langle I \rangle) \); thus, from (31), we again obtain the Poisson distribution \( P(m \mid \langle I \rangle) \), which is now independent of the field statistics. Observing the dependence of \( P(m) \) on \( A \) and \( T \) in the intermediate case, we may, in principle, obtain information on the scale of field coherence.

**Photon Bunching**

The relationship (31) between the distributions also determines the relations between the moments of photocounts

\[
\langle m^k \rangle \equiv \sum_{m=0}^{\infty} m^k P(m) \quad (35)
\]

and the moments of intensity

\[
\langle I^k \rangle \equiv \int_0^{\infty} dI \, I^k P(I). \quad (36)
\]

From (31), or immediately from (27), we find the relationship of the first moments:

\[
\langle m \rangle = T \langle W \rangle = \alpha \langle I \rangle. \quad (37)
\]

Using the method of generating functions (Section 3.4), we can easily find the following relationships between the variances as well:

\[
\Delta m^2 = \langle m \rangle + \alpha^2 \Delta I^2, \quad (38)
\]

where

\[
\Delta I^2 \equiv \langle I^2 \rangle - \langle I \rangle^2 \quad (39)
\]

and analogously for \( \Delta m^2 \). Thus, the photoelectron fluctuations, in addition to the usual Poisson (shot) part \( \langle m \rangle \), contain an additional contribution from light intensity fluctuations. Only in the case of a
single-mode stabilized laser \((\Delta I = 0)\) is this contribution absent. In other cases, electron fluctuations, in accordance with \((38)\), should, it would seem, always exceed the shot noise, since it follows from the definition \((39)\) and the condition \(P(I) \geq 0\) that \(\Delta I^2 \geq 0\).

The presence of these surplus fluctuations has come to be known as the \textit{photocount grouping} or \textit{bunching effect} (since the appearance of one pulse in the Poisson flux of pulses cannot, by definition, influence the appearance of the next, the inequality \(\Delta m^2 > \langle m \rangle\) signifies a "tendency" of pulses towards bunching). An analogous effect \(\Delta N^2 > \langle N \rangle\) for photon numbers is known as \textit{photon bunching} or \textit{correlation}. A similar effect was discovered by Brown and Twiss in 1956\textsuperscript{35} in the chaotic light of a mercury lamp.

It follows from \((11)\) and \((38)\) that, in chaotic light,

\[
\Delta m^2 = \langle m \rangle (1 + \langle m \rangle),
\]

\((40)\)
i.e., the excessive part of the variance is \(\langle m \rangle\) times greater than the Poisson part, with the result that the photon bunching effect is more clearly pronounced in classical fields. From a classical point of view, strong fluctuations of the wave amplitude \(|E_0|\) that are generated by a multiplicity of independent sources with random phases are to be expected. More unexpected for the classical theory is the \textit{antibunching} of photons and, accordingly, of photocounts, which contradicts \((38)\) and Mandel's initial formula \((31)\) if we assume that \(P(I) \geq 0\) in it (in the quantum theory, the last condition, as mentioned above, is violated).

\textbf{The Correlation of Intensities}

The distribution of photocounts \((31)\) does not provide direct information on either the temporal or spatial spectrum of radiation; this distribution is expressed through "one-point" correlation functions, with all the arguments being coincident (see \((32)\)). To determine fully the CF of the \(n\)th order, it is essential to measure the field at \(2n\) points of space-time. In the case of CF-1 this is done by means of interferometers, as schematically described above.

Let us now consider the measurement of the correlation function of
intensity, namely, the CF-2 of the following particular form:

\[ G^{(2)}(x_1, x_2; x_1, x_2) = G_{12}^{(2)}(r) = \langle I(x_1)I(x_2) \rangle \]

where \( x_i \equiv \{ x_1, t_i \}, r \equiv t_2 - t_1 \), and the vector \( r_2 - r_1 \equiv \rho \) is perpendicular to the direction of propagation. The colons are a reminder that, as we go over to the quantum theory, it is necessary, before averaging, to transpose all the operators \( E' \) to the right of \( E' \).

In the case of the Gaussian field, we find, with the help of (17),

\[ G_{12}^{(2)}(\tau) = \langle I(r_1)I(r_2) \rangle \left[ 1 + |g_{12}(\tau)|^2 \right], \tag{42} \]

where \( g_{12}(\tau) \) is the normalized CF-1. Thus, in the chaotic field, measuring the correlation of intensities provides information on the correlation of amplitudes. The relationship between \( G^{(2)} \) and \( G^{(1)} \) underlies light beating spectroscopy\(^{36}\) (the terms "the spectroscopy of intensity fluctuations" or the "proton correlation method" are also used).

In the general case, \( G^{(2)} \) is not expressed through \( G^{(1)} \) and the scale of second-order coherence may differ from \( \rho_{\text{coh}}, \tau_{\text{coh}} \) (thus, in two-photon light, sometimes \( \rho_{\text{coh}} \gg \rho_{\text{coh}}^{37} \)).

To measure the time dependence of \( G^{(2)}(\tau) \) at \( \rho = 0 \), a single detector with a delay line and an electronic correlator is used. Spectral analysis of photocurrent fluctuations may also be used. The first experiment of this type was carried out by Forrester, Hudmunsen, and Johnson in 1955, before the advent of lasers.\(^{38}\) State-of-the-art equipment makes it possible to obtain spectral resolution of a fraction of a Hertz.

To investigate spatial second-order coherence \( G^{(2)}(\rho) \), two detectors with a variable distance \( \rho \) between them should be used. According to (42), as \( \rho \) goes from \( \infty \) to 0, the photocurrent correlation doubles in the ideal case (Figure 1.17). This effect, which is sometimes called the correlation of intensities, was discovered by Brown and Twiss back in 1956\(^{35}\) and was subsequently used by them to measure the angular diameters of stars whose intensities correlate at distances on the order of hundreds of meters (instead of scores of meters, as in the case of Michelson's stellar interferometer).

Figure 1.18 shows a diagram of the Brown–Twiss experiment that measured \( G^{(2)} \) in the light of a mercury lamp. The splitting of the
beam by means of a semitransparent mirror makes it possible to measure the field correlation at any two points, however close. Let the photomultiplier operate as photon counter; then, by analogy with (27), the correlation of counts in the two channels $\langle m_1 m_2 \rangle$ is equal to $\alpha_1 \alpha_2 \langle I_1 I_2 \rangle$. From this we find, with the help of (42),

$$\langle m_1 m_2 \rangle = \langle m_1 \rangle \langle m_2 \rangle (1 + |g_{12}(0)|^2).$$  \hspace{1cm} (43)

This result is valid only in the case of single-mode detectors when the time constant of the detector $T$ is far lower than $\tau_{\text{coh}}$ (practically speaking, the width of radiation spectrum must be below $10^8$ Hz) and the detector's aperture $A$ far less than $A_{\text{coh}}$. If, on the other hand, $T \gg \tau_{\text{coh}}$, then a minor factor $\tau_{\text{coh}}/T$ appears in the second component of (43), which reduces the observable effect.

1.5 Multiphoton Effects: A Historic and Bibliographic Outline

The survey of the literature given below does not claim to be complete; it contains, for the most part, references to the earliest and most recent publications and reviews. Its aim is to give some background information on the history of research into spontaneous multiphoton effects.
Figure 1.18. A schematic of the first Brown and Twiss experiment to observe correlations of light intensity.

The Stone (Pre-Laser) Age

The first two-photon effect—the Rayleigh scattering of sunlight in the air—must have been observed by early man millions of years ago in the form of the blue color of the sky. It should be noted, however, that elastic Rayleigh light scattering is a linear effect. Photoluminescence, another long-familiar optical effect, can, in its simplest forms, be explained by the cascade three-photon process $\omega_3 \rightarrow \omega_2 + \omega_1$ (the $\hbar \omega_2$ energy is usually lost nonradiatively and the material heats up). Although the study of photoluminescence began centuries ago, Stoke’s law ($\omega_1 < \omega_3$) did not receive a satisfactory explanation until the arrival of the quantum theory and the concepts of the photon and energy levels.

In 1926, Vavilov and Levshin were the first to carry out experimental studies of the nonlinear optical phenomenon known today as the saturation effect; it is associated with a change in population levels (for references, see Reference 39). In 1928, Vavilov made a truly heroic attempt (considering the circumstances) to discover the violation of the superposition principle by intersecting light beams, i.e., in modern terminology, light-by-light scattering in vacuo. The subsequent experiments conducted by Vavilov and his associates in the 1930s, which involved the visual observation of light intensity fluctuations, are well known.

The first “genuine” two-photon nonlinear effect, Raman scattering, was discovered in 1929 simultaneously by Landsberg and Mandelshtam in the Soviet Union and by Raman and Krishnan in India (a detailed account of this discovery is contained in Reference 40).
Coherent scattering by thermal acoustic waves had been predicted by Brillouin (1922) and Mandelshtam (1926), and was discovered by Gross (1930). The quantum theory of this phenomenon was studied by Tamm in 1930.

In 1929, Mayer-Goppert examined the possibility of two-photon spontaneous radiation (for references to subsequent work in this area, see Reference 42). The simultaneous radiation of two optical photons was not experimentally discovered until 1965 by Lipeles et al., who used the photon count coincidence technique. The statistical properties of spontaneous two-photon fields were recently considered by Sokolov. Cascade two-photon transitions in which the nonsimultaneity of photon radiation in pairs is determined by the lifetime of the intermediate state have a far greater probability. Such transitions were observed in the optical range by Kaul in 1966 and were later used to set the limits of the semiclassical theory of radiation (for references, see References 46 and 47) and of the hidden variables theories and also for the absolute calibration of photodetectors. Two-photon transitions were discovered in the \( \gamma \)-range cascade back in 1947 by Brady and Deutsch and are now widely used in nuclear physics to investigate intermediate states (the \( \gamma \)-\( \gamma \) correlation method).

In the X-ray range, inelastic light scattering was first discovered as early as in 1923 by Compton. In 1934, Heitler and Nordheim calculated the probability of the double Compton effect, in which the pump photon turns into two photons upon interaction with the free electron. This process is analogous to three-photon Rayleigh scattering (or, if we allow for the recoil of an electron, to Raman scattering) and differs from parametric scattering in the incoherence of the contributions made by individual scattering particles. Coherent parametric scattering in the X-ray range was discovered in 1971 by Eisenberger and McCall.

In the opposite, radio frequency, region of the spectrum, multiphoton absorption on the rotational levels of gases was observed by Hughes and Grabner in 1950. A great variety of stimulated multiphoton effects in the radio frequency range was discovered shortly afterwards on the Zeeman sublevels in solid electronic paramagnetic substances. Due to low photon energy, other phenomena—such as Raman amplification, doubling and tripling of frequency, and five-quantum absorption—were observed in two-level spin systems.
Cascade frequency doubling\textsuperscript{60} and frequency subtraction\textsuperscript{61} were observed on three spin sublevels of the ground state of ruby. The "amplification upon saturation" effect was investigated in rotational and spin two-level systems.\textsuperscript{62–64} The corresponding spontaneous effects were later observed in the optical range.\textsuperscript{65–67}

The advent of paramagnetic and parametric UHF-range amplifiers in the 1950s raised the problem of determining their limiting sensitivity. The quantum noises of the simplest model of a parametric amplifier were studied by Louiseletal. in 1961\textsuperscript{68} (see also Reference 3). This work studied the change over time in the state of two modes of a cavity resonator in the case of the harmonic pump-induced modulation of the dielectric permeability of the medium filling the resonator. As a result, the following formula was obtained (cf. (1.1.39)):

\begin{equation}
N_1(t) = g(t)N_1(0) + f(t)[N_2(0) + 1],
\end{equation}

\begin{equation}
f(t) = g(t) - 1 = \sinh^2 \gamma t.
\end{equation}

The first experiments in which the correlation of light intensities (i.e., the fourth moment of the field or second-order coherence) played a role and which stimulated the progress of quantum optics were performed in the mid-1950s. Forrester et al.\textsuperscript{38} observed the subtraction of light frequency in the photoelectric effect in 1955.\textsuperscript{†} The following year, Brown and Twiss\textsuperscript{35} discovered the correlation of light intensities at two points in space. These phenomena formed the basis of a new trend in spectroscopy.\textsuperscript{36,69,70}

The Laser Age

In 1961, Franken et al.\textsuperscript{71} carried out the first experiment on light frequency doubling, thereby launching a new discipline, laser nonlinear optics. A large body of bibliographic literature is available on optic multiquantum transitions (e.g., References 7, 30, 31, 33, 34, 42, and 72–76) and in what follows we will be dwelling for the most part on works dealing with the radiation of biphotons.

In 1964, Robl\textsuperscript{19} investigated direct and cascade hyperparametric scattering in an optical nonlinear medium, proceeding from the effec-

\textsuperscript{†}The interpretation of this experiment is given in Reference 38a. The possibility of "heterodyning of light" was discussed by Horelick back in 1948.\textsuperscript{38b}
tive energy of perturbation $\chi^{(2)} E^3 + \chi^{(3)} E^4$. The “quantum noise” arising in a medium with modulated permeability as well as hyper-parametric scattering in a medium with cubic nonlinearity were investigated by Kroll in 1964. In 1966, Grinberg and Kramer drew an analogy between light-by-light scattering in a vacuum through the virtual creation of electron-positron pairs and light scattering in a solid body through virtual electrons and holes. As the width of the “forbidden zone” of the vacuum ($2mc^2 \sim 1$ Mev) is far greater than the forbidden zone of semiconductors ($\sim 1$ ev), a dramatic increase in the probability of four-photon processes in material should be expected over that in vacuum.

Three-Photon Parametric Scattering

The first numerical estimate of the intensity of three-photon parametric scattering in piezocrystals (Section 1.1), which was made by the author in 1967, yielded an unexpectedly high value with an effective temperature of $\sim 10^3$ K (see (1.1.13)), even with the use of a modest 1 w pump. The author was puzzled that this effect had not been detected earlier during experiments involving frequency subtraction and parametric amplification. None of the experimenters the author approached at the time believed in the reality of mysterious quantum noises when just those noises were discovered by scientists at Moscow University by accident in the course of experiments with a parametric light generator. In the same year (1967), parametric scattering was discovered independently by Harris et al. at Stanford University and by Magde and Mahr at Cornell University.

In the first experimental works, the radiation was interpreted not as the result of scattering but rather as the noise radiation of a parametric amplifier (it should be noted that here, too, the spontaneous effect was discovered later than the corresponding stimulated amplification process). However, if we exclude the case of megawatt pumps, then, according to the estimate (1.1.12), the actual amplification factor of the signal $\mathcal{G}$, allowing for inevitable losses, will be less than unity. It is therefore pointless to speak of “amplifier noises.” On the other hand, the analogy with Mandelshtam–Brillouin scattering by sound idler waves enables us to regard the parametric scattering effect as the result of scattering by light idler waves. It should be
noted that noncollinear scattering $(\theta_{1,2} \neq 0)$ with $\mathcal{G} \sim 1$ has the same intensity as collinear scattering.

The term “parametric scattering” was first used in Reference 82. Other authors used the terms parametric luminescence or fluorescence, optical parametric noise, parametric splitting of the light frequency, etc. It should be noted, however, that parametric scattering is no ordinary type of scattering; it can also be defined as the result of \textit{nonlinear diffraction}. Inelastic scattering implies an irreversible process accompanied by increasing entropy of the electromagnetic field, while parametric scattering may be described by an effective Hamiltonian (Chapter 6) that defines the reversible evolution of a closed system. In Reference 83, the possibility of partially restoring the coherent pump from the scattering field by means of a second piezocrystal is considered.\footnote{Such an experiment was recently performed by Abram et al.\textsuperscript{277}}

Subsequent experiments involving parametric scattering were described in References 84–122.\footnote{See also References 276–80.} Weinstein's work,\textsuperscript{97} in which a mercury lamp served as the pump, should be singled out for special mention. Chelma and Batifold\textsuperscript{116} observed scattering on the back-propagating idler wave. Burnham and Weinberg\textsuperscript{96} investigated the simultaneous radiation of signal and idler photons by the photon count coincidence method. In their series of works, Penin et al.\textsuperscript{93,107,112–114,117–121} used the parametric scattering and polariton scattering effects to determine the dispersion of the linear and quadratic polarizabilities of a number of crystals in the infrared, to measure the absorption coefficient, to analyze the nonstoichiometry of their composition, and also to investigate phase transitions and weak two-photon excitations; thus, we may now speak of parametric scattering spectroscopy as a new method of crystal spectroscopy. In the course of research by Krindach and this author,\textsuperscript{89} parametric scattering was detected visually and a connection was established between parametric and polariton scattering. These two types of scattering were observed simultaneously in Reference 93. Parametric superluminescence was observed in References 98, 103, 105, and 108.

The theory of parametric scattering was developed by the authors of References 123–36.\footnote{See also References 281–84.} A phenomenological description of both para-
metric and polariton scattering is given in References 93, 107, and 133. The influence of idler wave absorption was also investigated in References 125 and 130. Nonstationary parametric superluminescence was studied by Nazarova and the author.  

Field statistics in parametric scattering and the mutual correlation of signal and idler photons were studied by Zel'dovich et al. and Mollow. In Reference 135, it was proposed that this correlation be used for the development of standard light sources with known emission moments and a known number of photons, as well as for the absolute calibration of photomultiplier tubes (Section 6.4 and References 37 and 137–40). In References 135, 140, and 142, the possibility of using parametric scattering for the absolute measurement of the brightness of light is explored (Section 6.4).

The metrological potential of parametric scattering may form the basis of quantum photometry. In this connection, the problem arises of generalizing Kirchhoff's law (which underlies the existing standard of brightness) with reference to nonlinear optics (see Chapters 4–7 and References 29, 37, 138, 139, and 143). The associated problem of a nonlinear generalization of the fluctuation-dissipation theorem should be noted in this context (see Section 2.5 and References 144–151).

The authors of a number of works (see, e.g., References 152 and 153) have continued to investigate the time evolution of field statistics within the framework of the two-mode model put forward by Louisell et al.

Other Types of Scattering

Polariton scattering was first observed in a gallium phosphide crystal by Henry and Hopfield in 1965, two years before the discovery of parametric scattering. In 1966, during the study of polariton scattering in zinc oxide, an unsuccessful attempt was made to find polariton scattering in the upper branch of the dispersion curve (i.e., in our terminology, parametric scattering, which was not discovered in this crystal until 1975). A review of the extensive body of literature on polariton scattering will be found in References 156 and 157.

As noted in Reference 78, in addition to coherent parametric scattering through the macroscopic quadratic polarizability $\chi^2$, there should also occur an almost isotropic noncoherent scattering by den-
sity and orientation fluctuations of individual particles possessing quadratic hyperpolarizability \( \beta \). The theory of this three-photon Rayleigh or hyper-Rayleigh scattering by individual molecules or microheterogeneities (which resembles the double Compton effect\(^{53} \)) was investigated by Sokolovskii\(^{158} \), and by Baranova and Zel'dovich\(^{134} \). A relevant experiment is described in Reference 159. Asynchronous \((k_1 + k_2 - k_3 \neq 0)\) scattering is also possible in a uniform sample because of edge effects.\(^{124,134,160,161} \)

In a continuous medium with \( \chi^{(2)} = 0 \), it is possible to have three-photon Mandelshtam–Brillouin scattering in which the photon of the pump decays into two photons and an acoustic phonon of frequency \( \Omega \) and wave vector \( k \):

\[
\omega_3 = \omega_1 + \omega_2 + \Omega, \quad k_3 = k_1 + k_2 + K.
\]  

(2)

A corresponding stimulated process in which the acoustic wave is created by an external source was examined in Reference 25. It should be noted that, if we assume that \( \Omega = 0 \) in (2) and further assume that \( K \) stands for one of the vectors of the crystal's reciprocal lattice, then (2) will describe three-photon Bragg X-ray scattering or parametric scattering with a participating vector of the reciprocal lattice.\(^{54} \)

Simultaneously with the radiation of photons with frequencies \( \omega_3 \pm \Omega \) \((\Omega \ll \omega_1)\), the sample radiates photons with frequencies \( 2\omega_3 \pm \Omega \), which are near the frequency of the second harmonic of the pump (hyper-Rayleigh or hyper-Raman scattering, HRS). This phenomenon was predicted independently by Bersohn,\(^{162} \) Kielich,\(^{163} \) and Lee\(^{164} \) in 1964 and by Akhmanov and the author\(^{165} \) in 1965. It was observed by Maker et al.\(^{166} \) in 1965. Among the more recent works dealing with hyper-Raman scattering, see References 26 and 167–73, which contain more detailed references.\(^{\text{I}} \) Stimulated HRS has been observed by Dneprovskii et al.\(^{174} \) and Kaiser et al.\(^{175} \) (see also Reference 176).

Hyperparametric Scattering

Hyperparametric, or four-photon parametric, scattering (also called light-by-light scattering in matter), which is conveniently described by cubic polarizability,\(^{19,77,128,177} \) was first detected by Grinberg et

\(^{\text{I}}\)See also Reference 285.
al. in a cadmium sulfide crystal in 1968. Subsequent experiments are described in References 11–15, 178, and 179. The anti-Stokes quantum noises of frequency converters (i.e., cascade hyperparametric scattering in piezocrystals—see Section 7.1) were studied theoretically in References 19–24 and experimentally in References 16–18.

As mentioned in Section 1.3, when the frequency of the observed signal changes, hyperparametric scattering continuously changes into hyper-Raman scattering or into coherent Raman scattering (CRS), which was discovered by Terhune in the course of the first experimental studies of stimulated Raman scattering in the form of a directional cone of anti-Stokes radiation. The angles of preferred scattering of the anti-Stokes components in the case of a small divergence of the pump are in good agreement with the four-photon phase-matching condition (for references, see Reference 181—that work provides a good explanation of the deviations from ordinary phase-matching conditions that are sometimes observed; it also discusses the reasons for the nonisotropic nature of the higher components of coherent Raman scattering). Thus far, coherent Raman scattering has been observed in the stimulated mode with a powerful pulsed laser pump. In the spontaneous mode, the CRS effect is quadratic with respect to the pump and is due to the quantum fluctuations of the Stokes field, as well as to the thermal or quantum fluctuations of molecules (Section 7.2). In Reference 29, the present author demonstrated theoretically that spontaneous coherent Raman scattering is accompanied by biphoton radiation in the case of a low sample temperature (Section 7.3). The correlation of intensity in the case of the scattering of a standing-wave pump wave was also considered in References 182–84. The correlation of photons of different frequencies should occur in resonance fluorescence.

Spontaneous coherent Raman scattering has a corresponding stimulated effect of resonance frequency conversion, $2\omega_3 - \omega_2 \rightarrow \omega_1$, $\omega_3 - \omega_1 \sim \omega_\mu$, which has found important spectroscopic applications (the active spectroscopy technique, or CARS). This method was first used by Maker and Terhune in 1965 (see Reference 27). A phenomenological description of CARS of polaritons is given in Reference 187.

In 1974, Harris and Miles proposed using another variant of resonance frequency conversion, viz., two-photon pump resonance with
an electron transition $2\omega_3 \pm \omega_2 \rightarrow \omega_1$, $2\omega_3 \sim \omega_\mu$, for the generation of ultraviolet (UV) radiation (for details, see References 31, 34, and 42). This stimulated effect had been discovered earlier in the radio frequency range.\(^{58}\) No corresponding spontaneous process (Figure 1.9k), which might be described as two-photon resonance fluorescence, has been observed to date.

Finally, resonance fluorescence occurs in one-photon pump resonance with an electron transition $\omega_3 \sim \omega_\mu$; this was first investigated by Wood way back in 1905. In the higher approximations with respect to pump intensity, one has to allow for saturation effects, resonance splitting, and hyper-Raman scattering on the electron transition $\omega_1 = 2\omega_3 - \omega_\mu$ (Figure 1.9j) or, which amounts to the same thing, resonance hyperparametric scattering. These effects were first studied in 1961 by Rautian and Sobelman\(^{189}\) and in 1968 by Newstein.\(^{190}\) The first experimental studies were carried out by Schuda et al.\(^{65}\) For references to subsequent work and for a more detailed description, the reader is referred to References 30, 31, 34, 42, 66, 67, 190, and 191.\(^{**}\) The corresponding stimulated effect was investigated in the radio frequency range earlier in the 1960s.\(^{62-64}\)

Quantum Optics

A traditional category of prelaser statistical optics, the coherence theory of the first order was based on classical electrodynamics. The first experiments involving the observation of second-order coherence were performed between 1955 and 1956 and also admitted of a classical interpretation. In 1958, Mandel,\(^{193}\) proceeding from the semiclassical theory, obtained a formula that related field statistics to the statistics of the observed parameter, the number of photocounts at the output of the photomultiplier tube (Sections 1.4 and 3.4).

Quantum statistical optics entered a period of intensive development during the 1960s, after the advent of the laser and nonlinear optics. An extensive bibliography is contained in Reference 8\(^{**}\) and here we will mention only a few selected works.

A major landmark in the history of quantum optics was reached in 1963 when Glauber evolved a technique for describing a field in

\(^{**}\)See also Mollow’s review article.\(^{286}\)

\(^{**}\)See also the later publications, References 287 and 288.
terms of coherent states and the definition of the hierarchy of normal correlation functions. Glauber–Sudarshan’s $P$ representation for the density matrix of the field (see Section 3.3) made it possible to compare quantum and classical statistics more directly. We will note recent generalizations of the $P$ representation and Mandel's formulas that cover, in particular, squeezed states of the field.

The basic subject matter of quantum optics is light with extraordinary, i.e., non-Gaussian, statistics, notably coherent light emitted by an ideal laser. In this case, the field is, from a classical standpoint, totally determinate and is in a coherent state from the quantum point of view. Since the mid-1960s there has been a substantial and growing number of publications dealing with the semiclassical and fully quantum statistics of laser emission (see References 1–6, 195, and 196). We shall mention only one of the more recent theoretical works, according to which a slight antibunching should occur in laser light.

The states of the field that have no classical analogues are, without doubt, of considerable interest to students of quantum optics. From the standpoint of information theory and the problem of gravitational wave detection, squeezed states are of interest. The possibility that such states exist was first noted back in 1965 by Robinson and Takahashi. The properties and possible techniques for the preparation of this class of states were subsequently examined by Yuen and by many other workers (see, e.g., References 204–9). There has, as yet, been no experimental observation of such states.

States of the field involving photon antibunching were first referred to by Glauber in his 1963 lectures. A variety of states having this property have been studied by the authors of References 210–33 (see Paul’s review article). Antibunching was discovered experimentally in 1977, when Kimble, Dagenais, and Mandel detected it in the light of resonance fluorescence.

Superbunching (or enhanced bunching) arises, for instance, (a) during the spontaneous two-photon decay of excited atomic levels (this was discovered and commented upon in 1965); (b) in the case of parametric scattering or hyperparametric scattering (predicted in 1969 and discovered in 1970), (c) in the case of Raman scattering (see also References 20, 138, 139).
Chapter 7), and (d) in the case of resonance fluorescence.\textsuperscript{182,184,185}

So far, superbunching has been detected at two different points of the field with the help of two photomultiplier tubes and a coincidence circuit. "One-point" superbunching was detected recently with the help of one photomultiplier and used for the absolute calibration of photomultiplier tubes.\textsuperscript{236}\textsuperscript{55} The present author noted the practical possibility that "transverse" superbunching might be observed on the display screen of an electronic image converter and that its quantum efficiency might be measured by means of parametric scattering in Reference 37. That work also contains perhaps the first discussion of the impact of field statistics on the photographic process.

It should be emphasized that, in experiment, light with non-Gaussian statistics arises due to the optical anharmonism of matter. Thus, saturation in a single-mode laser results in a coherent state of the field. Generally speaking, two-level anharmonism will, under a variety of conditions, produce squeezed states,\textsuperscript{208} antibunching states,\textsuperscript{32} or superbunching states.\textsuperscript{211}

Cubic anharmonism describes spontaneous and stimulated two-photon transitions and leads, as a result, to superbunching (Sections 5.4 and 7.3) and to antibunching.\textsuperscript{6,222,223} A two-photon laser (see the references in References 238 and 239) proposed in 1964\textsuperscript{240,241} produces, according to Reference 202, a squeezed state, (see, however, References 242–46).

The interaction of three modes due to quadratic susceptibility results in antibunching,\textsuperscript{212,214} superbunching (Section 6.4 and References 37 and 96, squeezed states\textsuperscript{206,207} and states involving odd moments (Section 5.3 and References 8, 138, 139, and 143).

\textsuperscript{55}See also Reference 277.
References


401
REFERENCES

77. Kroll, N. in Reference 1, p. 9.


REFERENCES

REFERENCES