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Non linear optics

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INTRODUCTION

1.1 BRIEF HISTORY

What is light? What is it made up of? How is it generated? How fast does it travel? How does it propagate across empty space? How does it behave when it comes across an object? How does it interact with matter? These are some of the many questions that arise in our mind. Optics is the branch of physics, which deals with such questions and describes about the phenomena and laws associated with the generation, and propagation of light and its interaction with matter. We briefly answer these questions in this introductory chapter in the evolution of our understanding about light.

A) Development of Geometric Optics:

The Greeks were aware of the rectilinear propagation of light. They knew that when light is reflected from a mirror, the angle of incidence is equal to the angle of reflection. This was stated by Euclid (300 B.C.) in his book Catoptrics. Hero of Alexandria suggested that light traverses the shortest path between two points. They were also aware of refraction of light as it passes from one transparent medium to another. Claudius Ptolemy (130 A.D.) of Alexandria measured the angles of incidence and refraction for several media. Further progress came to a halt with the fall of Roman Empire in 475 A.D. Study of light was again revived in Europe during the thirteenth century. Francis Bacon (1215-1294) suggested the idea of using lenses to improve eyesight. In about 1280, spectacle lenses came into use to correct faulty vision. In 1609 Galileo (1564-1642) devised a practical telescope. Van Leeuwenhoek (1632-1723) developed the first microscope. John-Kepler discovered the phenomenon of total internal reflection. In 1621 Willebrod Snell (1591- 1626) and independently in 1637 Rene Descartes (1596-1650) discovered the law of refraction. In 1658, Fermat (1601-1655) discovered the principle in 1609, Galileo devised a of least time. According to this principle, light always follows that path practical telescope. This takes it to its destination in the shortest time. He re-derived the law of reflection and refraction applying this principle of least time for the path followed by light. In 1660 the phenomenon of diffraction was noticed by Grimaldi (1618-1663). In 1667 Newton established that white light is composed of seven independent colours. In 1670, Bartholinus (1625-1698) discovered the phenomenon of double refraction.

In 1675 Isaac Newton (1642-1727) put forward the corpuscular theory. According to this theory, a luminous body emits in all directions streams of extremely minute particles, called corpuscles. They are supposed to travel through a medium with a tremendous but finite velocity in straight line paths. The particle theory of Newton could explain the straight line propagation of light and that an object casts a sharp shadow; but it failed to explain why the continued loss of particles did not also cause a source of light to lose weight. However, the theory could prove the laws of reflection and refraction of light. Newton predicted that light should travel faster in a denser medium than in a rarer medium. However, the phenomenon of diffraction and Newton's rings could not be explained on the basis of corpuscular theory.

In 1676, Romer (1644-1710) proved that light travels with a finite velocity. Robert Hooke (1635-1703) studied the coloured patterns formed due to thin film interference.

B) Development of Wave Optics:

In 1678 Huygens (1629-1695), a contemporary of Newton, proposed wave theory of light. According to this theory, light energy is supposed to be transferred from one point to another in the form of waves. Huygens was able to prove the ordinary laws of reflection and refraction. He predicted that light should travel slower in a denser medium than in a rarer medium. He also explained the phenomenon of double refraction by assuming two types of waves. The wave theory was not accepted immediately. The chief reason was that a wave motion needs a medium; but light could travel to us from the sun through the vacuum of space.

In 1803, Thomas Young (1773-1829) demonstrated for the first time the interference of light beams.He also explained Newton's rings and the colours of thin flims on the basis of interference of light waves. Thomas Young provided strong support to the wave theory. In 1808, Malus (1775-1812) discovered the polarization of light. In 1815, Augustin Fresnel (1788-1827) further developed the wave theory and explained the rectilinear propagation of light which has been the chief obstacle in the way of accepting wave theory. He provided a satisfactory explanation of the diffraction phenomenon.

Following Huygens, both Young and Fresnel assumed that light waves are longitudinal. Young and Fresnel conceived of an elastic medium, which was assumed to exist pervading the entire universe, and it was named luminiferous ether. The vibrations of the ether propagated as light, just as longitudinal vibrations in air propagate as sound. But the longitudinal wave theory of light could not explain polarization, a property exhibited by transverse waves but not by longitudinal waves. Fresnel and Arago (1786-1853) conducted experiments on superposition of linearly polarized light. Young eventually realized that light is a transverse wave and in1817 explained the results of Fresnel and Arago's experiments. In 1850, Jean Foucault (1791-1868) established that light travels slower in liquids than in air. This is just opposite to the prediction of Newton's theory.

Finally, the wave model was accepted. The acceptance of the wave theory of light made it obvious that a supporting medium should exist. Subsequently, elastic ether theory was developed during the next ten years. Strange properties were attributed to it. It was assumed to be extremely rigid so that it can support the exceedingly high frequency oscillations of light travelling at a speed of 3×10^8 m/s; yet it does not offer resistance to the motion of celestial bodies through it. Its density was supposed to increase in material substances to account for the lower velocity. In 1823, Fresnel derived expressions for the reflection and transmission coefficients on the basis of ether theory.

C) Nature of light:

Around1836, Faraday (1791-1867) showed that a varying magnetic field induces an electromotive force and thus established the intimate connection between electricity and magnetism. Further, Faraday showed that the polarization of light was affected by a strong magnetic field, which was the first hint as to the electromagnetic nature of light. Clerk Maxwell (1831-1879) unified the empirical laws of electricity and magnetism into a coherent theory of electromagnetism. In 1873, Maxwell showed that the speed of electromagnetic waves equals the speed of light. On the strength of this, he made the prediction that light is a high frequency electromagnetic wave. In 1887, Hertz (1857 -1894) confirmed Maxwell’s theoretical prediction by producing and detecting electromagnetic waves. The electromagnetic waves were initially supposed to be supported by the ether medium. Though electromagnetic theory is capable of explaining the phenomena connected with the propagation of light, it fails to explain the processes of emission and absorption. H.A.Lorentz (1853-1928) assumed that ether is in a state of absolute rest to be the carrier of electromagnetic field.

In 1887, Michelson-Morley performed the famous ether-drift experiment and found that light travels at the same speed irrespective of the position of the earth in its orbit. It led to the conclusion that ether does not exist. Hence, light is a self-sustaining high frequency electromagnetic wave. This theory is known as the Field Theory.

D) Development of Quantum Optics:

In 1814 Fraunhofer discovered dark lines in the solar spectrum. In 1861 Bunsen and Kirchhoff attributed them to the absorption of certain wavelengths by the gases in the outer atmosphere of the sun. It was also found that every gaseous chemical element possesses a characteristic line spectrum. The detailed studies of emission and absorption spectra of elements evolved into a separate discipline. In 1900, in order to obtain a correct theoretical expression for the black body radiation, Max Planck (1858-1947) found it necessary to suppose that light is absorbed or emitted in the form of elementary quanta. In 1905, Einstein (1879-1955) made use of the quantum concept to successfully explain the photoelectric emission. According to him, light is a stream of photons. In 1913, applying Planck's quantum hypothesis, Niels Bohr (1885-1962) devised an atomic model for the emission and absorption of light. It successfully explained the simple laws of line spectra of gases.The traditional sources of light produce incoherent light.The first coherent source of light, namely laser was built in 1960.The high power lasers led to a number of nonlinear optical effects such as harmonic generation, frequency mixing etc. Quick developments in holography and fibre optics followed the discovery of lasers.

We now visualize a photon as a bundle of electromagnetic radiation that oscillates with a definite frequency and travels through free space with the speed of light. Individual photons carry energy and momentum, so light has particle-like properties. When the number of identical photons is very large, they exhibit the properties of a continuous wave with the same definite frequency and propagation speed as the quantum. The phenomena of interference, diffraction and polarization and propagation of light in space is adequately explained by classical electromagnetic wave theory, whereas the experiments involving interaction of light with matter, such as photoelectric effect ere best explained by assuming that light is a particle.

1.2 THE FOUR IMPORTANT THEORIES

Various theories have been put forward about the nature of light. We will make a brief survey of the four important theories which guided the evolution of our understanding of the nature of light. The theories are known as

1. Corpuscular theory
2. Wave theory
3. Electromagnetic theory and
4. Quantum theory.

1. CORPUSCULAR THEORY

The corpuscular theory was postulated by ancient Greeks and was favoured by Sir Isaac Newton. According to this theory, a luminous body continuously emits tiny, light and elastic particles called corpuscles in all directions. These particles or corpuscles are so small that they can readily travel through the interstices of the particles of matter with the velocity of light and they possess the property of reflection from a polished surface or transmission through a transparent medium. When these particles fall on the retina of the eye, they produce the sensation of vision. On the basis of this theory, phenomena like rectilinear propagation, reflection and refraction could be accounted for, satisfactorily. Since the particles are emitted with high speed from a luminous body, they, in the absence of other forces, travel in straight lines according to Newton's second law of motion. This explains rectilinear propagation of light.

2. WAVE THEORY

The test and completeness of any theory consists in its ability to explain the known experimental facts, with a minimum number of hypotheses. From this point of view, the corpuscular theory is above all prejudices and with its help rectilinear propagation, reflection and refraction could be explained. By about the middle of the seventeenth century, while the corpuscular theory was accepted, the idea that light might be some sort of wave motion had begun to gain ground. In 1679, Christian Huygens proposed the wave theory of light. According to this, a luminous body is a source of disturbance in hypothetical medium called ether. This medium pervades all space. The disturbance from the source is propagated in the form of waves through space and the energy is distributed equally, in all directions. When these waves carrying energy are incident on the eye, the optic nerves are excited and the sensation of vision is produced. These vibrations in the hypothetical medium according to Huygens are similar to those produced in solids and liquids. They are of a mechanical nature. The hypothetical ether medium is attributed to the property of transmitting elastic waves, which we perceive as light. Huygens assumed these waves to be longitudinal, in which the vibration of the particles is parallel to the direction of propagation of the wave.

Assuming that energy is transmitted in the form of waves, Huygens could satisfactorily explain reflection, refraction and double refraction noticed in crystals like quartz or calcite. However, the phenomenon of polarization discovered by him could not be explained. It was difficult to conceive unsymmetrical behaviour of longitudinal waves about the axis of propagation. Rectilinear propagation of light also could not be explained on the basis of wave theory. The difficulties mentioned above were overcome, when Fresnel and Young suggested that light wave is transverse and not longitudinal as suggested by Huygens. In a transverse wave, the vibrations of the ether particles take place in a direction perpendicular to the direction of propagation. Fresnel could also explain successfully the rectilinear propagation of light by combining the effect of all the secondary waves starting from the different points of a primary wave front.

3. ELECTROMAGNETIC THEORY

In 1862 Maxwell ingeniously synthesized electricity and magnetism and developed equations which succinctly combine the important theories. He showed that electromagnetic waves travel with the speed of light and hence drawn the most important conclusion that light wave itself is an electromagnetic wave. Initially, the existence of ether medium was presumed from propagation of electromagnetic waves in space. However, if light waves which are of very high frequency are to propagate, and at the same time allow a free passage to heavenly bodies, then the ether have to be rigid as well as pliable. It became impossible to visualize the hypothetical solid which could be easily compressed or extended, could permit resistance-free passage of heavenly bodies through it, and yet be elastic to twisting or bending stresses in order to allow propagation of waves. Ultimately, in 1887 Michelson and Morley proved conclusively that there was no ether surrounding the earth or elsewhere.

4. QUANTUM THEORY

While experimenting on the black body radiation, Max Planck had come to the conclusion that the absorption or radiation of energy is not a continuous process. He postulated that thermal radiation is emitted or absorbed intermittently by indivisible amounts of energy called quantum. Each quantum carries an energy hv where h is a constant now called Planck's constant. Einstein elaborated the quantum concept in an endeavour to account for the phenomenon of photoelectric emission. He postulated that the quanta travel in space as separate entities with the speed 'c'. The quanta are named as photons. The further confirmation for the quantum theory is obtained when Compton Effect was discovered in 1923. Compton found that when monochromatic x-rays fell upon matter, the scattered rays contained not only the original x-rays but also x-rays of wavelengths longer than the original. Though the quantum theory explains successfully the interaction of radiation with matter, it cannot account for the phenomenon of polarization, interference and diffraction. The contradictory aspects are reconciled by postulating dual nature of radiation. Accordingly, radiation is viewed as having both the particle as well as wave nature.

1.3 INTRODUCTION OF NONLINEAR OPTICS

Lasers generate coherent radiation at many wavelengths ranging from meter wavelength region to the soft x-rays region. However, it is not possible to produce light covering all wavelengths of interest in spite of the fact that a large number of active materials are available and lasers can be built using them. It becomes, therefore, necessary to transform the frequency of light generated by lasers into light of desired frequency. Nonlinear optical media help us generate frequencies that were not available, through frequency conversion techniques. Harmonic generation, sum and difference frequency generation and parametric oscillations are some of the important nonlinear processes utilized in laser light frequency transformations. Stimulated Raman scattering is another important process that is used in generating new wavelengths. The processes such as second harmonic generation, sum and difference frequency conversion, parametric oscillation are associated with passive media, i.e., media that do not make evident their own characteristic frequencies. Stimulated Raman scattering arises in active media that impose their characteristic frequencies on the light wave. The beginning of the field of nonlinear optics is often taken to be the discovery of second-harmonic generation by Franken et al. (1961), shortly after the demonstration of the first working laser by Maiman in 1960.

1.4 A BRIEF HISTORY OF NONLINEAR OPTICS

You are about to embark on a study of nonlinear optics. Before we delve into the mathematical framework behind nonlinear optics, it is instructive to briefly present an historical perspective. The field of nonlinear optics encompasses a rich diversity of phenomena whose applications are growing at a seemingly exponential rate. The modern rebirth of the field began with the proposal of a laser device in 1958 by Arthur L. Schawlow and Charles H. Townes and its first operational demonstration in 1960 by Theodore Maiman. The laser was conceived as a highly monochromatic and coherent light source with emission at “optical” wavelengths, that is, at wavelengths far shorter than maser wavelengths. Indeed, the laser was initially so closely tied to the development of the maser, which generates coherent radiation at microwave frequencies, that in early publications it was referred to as the *optical maser.*

In the nineteenth century, there were two noteworthy discoveries of field-induced refractive birefringence effects. A quadratic field effect was described by the Scotsman John Kerr in 1875 and a linear field effect was reported by the German Friedrich C. A. Pockels in 1883. Today we recognize these effects as third- and second-order nonlinear effects, respectively. In both cases, these phenomena affect the phase of the wave, rather than the amplitude. In 1922, Leon N. Brillouin, a French physicist, postulated a nonlinear coupling between acoustic and optical waves in a medium, and in 1928, two Indian physicists Chandrasekhara V. Raman and Kariamanickam S. Krishnan measured an inelastic spontaneous emission effect, now commonly called the Raman effect, which was predicted in 1923 by the Austrian Adolf Smekel. Both of these effects are third-order nonlinear phenomena. Interestingly, the Russian physicist Leonid I. Mandelstam independently predicted both of these effects during that period.

Two publications of note in the history of nonlinear optics appeared in the following decade. In 1931, the German Maria Goepport-Mayer published her PhD dissertation work on two-photon absorption processes by extending concepts of quantum theory. Two-photon absorption is also a third-order nonlinear effect that corresponds to an imaginary contribution to the coefficient, whereas the Kerr effect corresponds to a real third-order contribution. The effect was observed soon after the invention of the laser as a nonlinear fluorescence phenomenon. Her work became honored by coining a molecular cross section of a Goeppert-Mayer (GM), which has the unit 10^{-50} cm^4 s/photon; it is a convenient unit in studies of nonlinear absorption in mate- rials. The second publication is the paper by Albert Einstein, Boris Podolsky, and Nathan Rosen in 1935 (at that time at Princeton University) that purported to call into question the inadequacy of quantum mechanics. In their argument, they introduced what is now called quantum entanglement of particle pairs that obey momentum and energy conservation laws. This has been a fruitful field of research in nonlinear optics by using parametric down-conversion to split a photon into a pair of photons. This research is leading to the emergence of new fields in quantum information and quantum communications.

Within a few years after the laser was invented, there was an explosion of laser-related activities around the globe. Within a few years, nonlinear optical effects such as second-harmonic generation (SHG), sum-frequency generation (SFG), difference-frequency generation (DFG), optical rectification, two-photon absorption, and thirdharmonic generation were observed for the first time at optical frequencies (see Figure 1.1 and references therein). The celebrated paper in 1961 by Peter Franken's group at the University of Michigan reported a first measurement of SHG using a quartz crystal as the nonlinear material. The published paper is also interesting for what it does not show, namely, the second-harmonic signal was interpreted by the copy editor as a smudge on the photograph and the proof of SHG signal was removed. The paper caused quite a stir because it provided new insights into optical frequency conversion at a quantum mechanical level; the new conceptualization of nonlinear optical processes was not widely appreciated at the time.

Additional studies quickly followed building a theoretical framework to describe and predict the observed nonlinear behavior. Nikolaas Bloembergen's group at Harvard University was among the early contributors (Armstrong et al., 1962; Bloembergen and Pershan, 1962; Kleinman, 1962a,b; Pershan, 1963; Ducuing and Bloembergen, 1964; Harris, 1966; Minck et al., 1966; Tang, 1966; Boyd and Kleinman, 1968; Giallorenzi and Tang, 1968). The landmark 1962 paper by Armstrong et al. presented analytical solutions of nonlinear equations and proposed new experimental strategies to achieve efficient nonlinear conversion. The invention of the laser ushered in an era of publications reporting observations of new phenomena, exploring new aspects of the field's theoretical underpinnings, and improving upon earlier results. Advances in laser and photodetection technologies have enabled studies of additional nonlinear phenomena in extended wavelength regimes and the creation of ultrashort pulses.

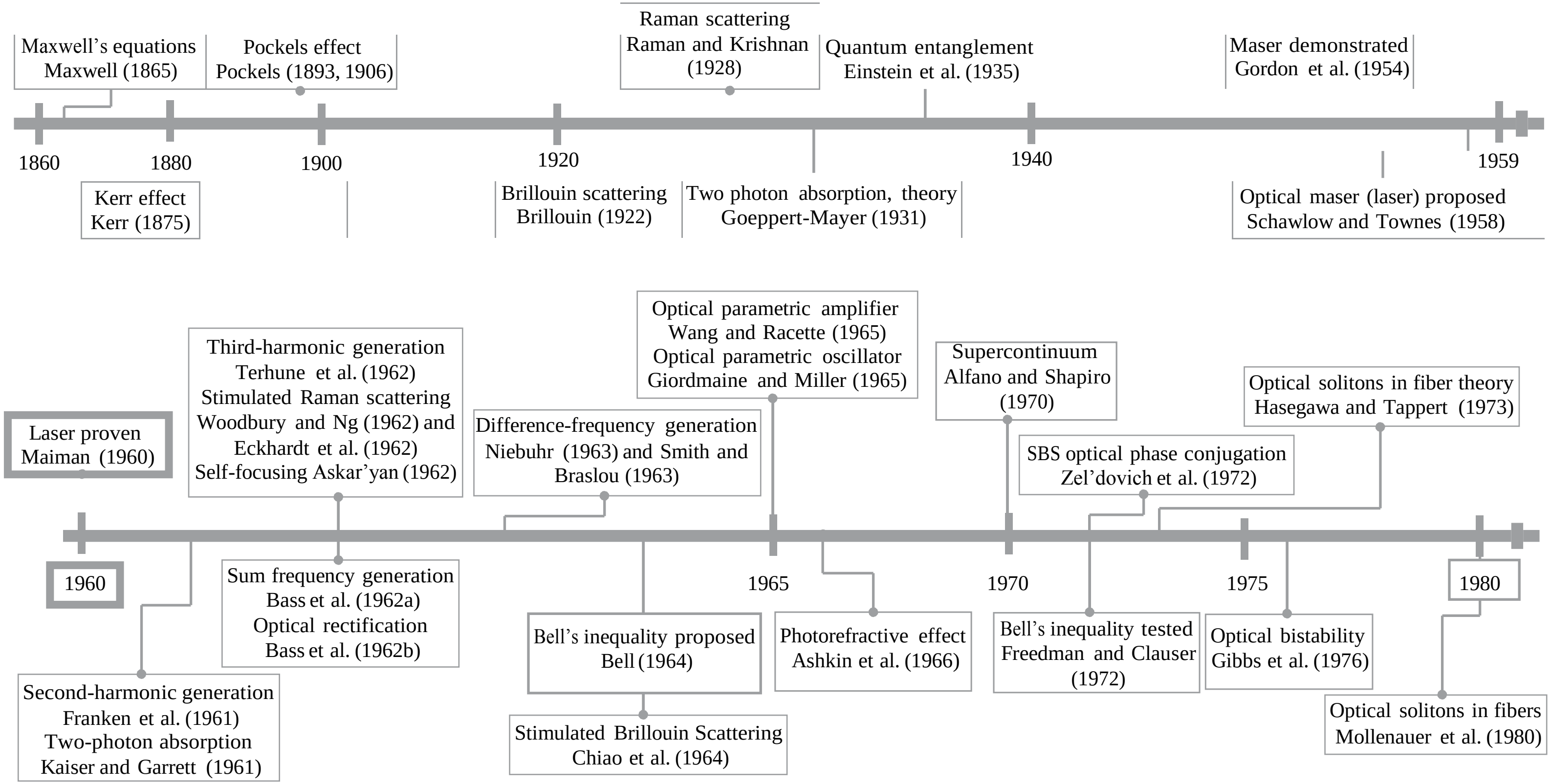

FIGURE 1.1 Timeline pointing out selected milestones in the history of nonlinear optics. *Top*: Prehistory of nonlinear optics before the laser's invention. *Bottom*: A 20-year span of rapid nonlinear optics development after the laser was demonstrated.

Behind all these developments is the high field associated with lasers. Because of their temporal and spatial coherence, lasers have brightness, defined as W/m^2/Sr, which far exceeds other light sources. Early *Q*-switched lasers easily attained intensities of >1 MW/cm^2 by focusing, and the interaction of these high fields with materials led to observations of nonlinear behavior. The rate of discovery and development in laser science and in nonlinear optics since the early 1960s continued at a rapid pace in subsequent decades and was highly synergistic. The generation of ultra-short pulses requires nonlinear elements to mode-locked pulses in the cavity. Laser operation down to the femtosecond pulse width range is now a common tool in the lab and in commercial machining applications. The development of super-intense femtosecond lasers opened a new regime of strong-field nonlinear optics. Strong fields drive a nonlinear response in gases that can generate harmonic waves with over 100 times the energy of the pump photons; these phenomena are called high-order harmonic waves and photon wavelengths can reach into the x-ray regime. Due to the coher- ence of the pump laser source, the high-order harmonics also have some measure of coherence; coherence combined with the shorter wavelengths has also lead the way to breaking the femtosecond barrier and producing attosecond pulse widths.

From among the many different nonlinear phenomena reported, we mention here several of those covered in this book: spontaneous parametric fluorescence in 1967 (Akhmanov et al., 1967; Byer and Harris, 1968), optical phase conjugation in 1972 (Zel'dovich et al., 1972; Yariv, 1976), optical bistability in 1976 (Gibbs et al., 1976), and optical solitons in 1980 (Mollenauer et al., 1980).

Applications for nonlinear optical devices were readily apparent after the underlying nonlinear effects were first demonstrated. For instance, the ability to convert laser light to new frequencies adds significant flexibility to fixed frequency laser sources. Moreover, the efficiency of nonlinear frequency conversion can be high, in some cases depleting the input laser energy. It is common to find commercial lasers sold with frequency extension units to double, triple, and quadruple the fundamental frequency. Continuously tuned nonlinear sources are the result of parametric frequency conversion processes. Frequency conversion is more than a convenience nonlinear optical techniques enable experiments that require specific frequencies unattainable by available lasers, which have limited tuning ranges.

Another class of nonlinear optics applications called nonlinear spectroscopy has established an understanding of molecular materials at the quantum level. Nonlinear absorption and scattering methods provide a means to identify trace quantities of unknown material constituents with high fidelity. Spontaneous Raman scattering (Raman and Krishnan, 1928) imparts a “fingerprint” scattered frequency signature that allows for the identification of different materials. The increased brightness of laser beams has rendered stimulated Raman scattering (SRS) signals observable and using a laser beam incident on a suitably prepared surface SRS spectroscopy has improved molecular fingerprinting sensitivity by 10–12 orders of magnitude, even enabling single-molecule detection. Nonlinear saturation spectroscopy allows for high-resolution spectroscopy by eliminating Doppler shifts in absorption features, called Lamb-dip spectroscopy (Lamb, 1964). Many laser-based spectroscopy topics are not treated in this book; the interested reader may wish to consult a survey of spectroscopic techniques for further information (Demtroder, 1996).

1.5 UNITS

Nonlinear interactions are described using a variety of systems of units in the literature. The two most common ones are SI and Gaussian (cgs, esu) units. Gaussian units are commonly referred to as “esu,” shorthand for “electrostatic units,” in the literature. Each system has its advantages, but we adopt SI for all formulae. Formulae written in SI units are particularly helpful when working with experimentally measured values. It is useful to relate the SI and cgs values of certain material properties.

$$\chi^{(1)}_{\mathrm{SI}} = 4\pi\chi^{(1)}_{\mathrm{Gaussian}},$$

$$\chi^{(2)}_{\mathrm{SI}} = \frac{4\pi}{3\times10^{4}}\chi^{(2)}_{\mathrm{Gaussian}},$$

$$\chi^{(3)}_{\mathrm{SI}} = \frac{4\pi}{(3\times10^{4})^{2}}\chi^{(3)}_{\mathrm{Gaussian}}.$$

The denominator is the conversion factor from stat volts/cm to volts/m and the factor of 4π in the numerator is a notational change in the definition of the susceptibilities. The form of Maxwell’s equations in Gaussian units has a factor of 4π multiplying the source terms and a factor 1/c multiplying the flux contributions in both Faraday’s and Ampere’s laws, which means that electric and magnetic fields have the same units.

1.5 SOME MATHEMATICAL TOOLS

There are a few linear transformations often encountered in optics, which transform a function into another function (e.g. the Fourier transformation) or into a number (e.g. the Dirac delta). We review these basic operations, also in order to clarify notational conventions.

The Fourier transform of a function $f(t)$ is given by:

$$F_\omega[f(t)] = f(\omega) := \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}t\ \ f(t)\mathrm{e}^{-\mathrm{i}\omega t} \tag{1.1}$$

In solving differential equations, such as the wave equation, it is often helpful to use Fourier transformation. This explains the importance of this mathematical tool in optics. The inverse Fourier transform is:

$$F_t^{-1}[f(\omega)] = f(t) = \int_{-\infty}^{\infty} \mathrm{d}\omega\ \ f(\omega)\mathrm{e}^{\mathrm{i}\omega t} \tag{1.2}$$

It can be interpreted as the composition of the signal $f(t)$ from harmonic basis functions (Fourier components), with (complex) amplitudes $f(\omega)$. We note that a time-periodic function has discrete Fourier component spectrum.

The Dirac delta is defined as follows:

$$D_{t_0}[f(t)] := f(t_0) \tag{1.3}$$

It is usually written in an integral form with the help of the Dirac delta ‘function’:

$$\int_{-\infty}^{\infty} \mathrm{d}t\ \ \delta(t-t_0)f(t) := D_{t_0}[f(t)] := f(t_0) \tag{1.4}$$

It holds $\delta(t) = \delta(-t)$. It is also visible from the integral form that the delta function has the inverse dimension of its argument: $[\delta(t)] = 1/[t]$. For the Dirac delta function, mathematically being in fact a distribution rather than a function, the following Fourier representation is also useful:

$$\delta(t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\omega \mathrm{e}^{\mathrm{i}\omega t} = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}\omega \mathrm{e}^{-\mathrm{i}\omega t} \tag{1.5}$$