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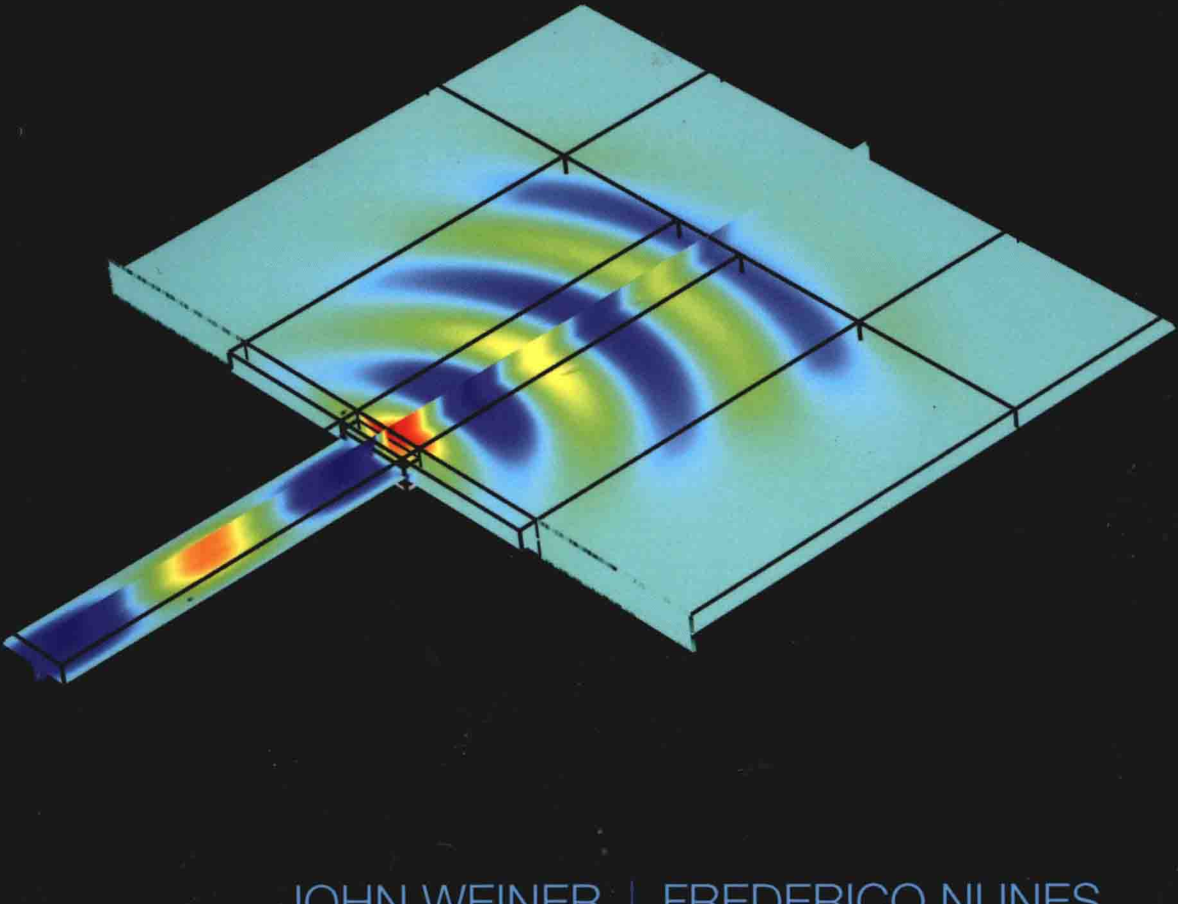
Light–Matter Interaction

Physics and Engineering at the Nanoscale



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Physics and Engineering at the Nanoscale

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To Annick and Marize for patience and encouragement.

Preface

Light–matter interaction is pervasive throughout the disciplines of optical and atomic physics, condensed matter physics, and electrical engineering with frequency and length scales extending over many orders of magnitude. Deep earth and sea communications use frequencies of a few tens of hertz and X-ray imaging requires sources oscillating at hundreds of petahertz ($10^{15}\,\mathrm{s}^{-1}$). Length scales range from thousands of kilometers to a few hundred picometers. Although we cannot pretend to offer an exhaustive treatise on this vast subject over all these frequency and length scales, we do propose to provide advanced undergraduates, graduate students and researchers from diverse disciplines the principal tools required to understand and contribute to rapidly advancing developments in light–matter interaction centered at optical frequencies and length scales around a hundred nanometers.

After a historical synopsis of the principal ideas leading to our present understanding of light and matter in Chapter 1, we enter the heart of the subject with a review of classical electrodynamics in Chapter 2. The intent is to reacquaint the reader with electric and magnetic force fields and their interactions with ponderable media through Maxwell's equations and the Lorentz force law. We emphasize here macroscopic quantities of permittivity and permeability and, through the constitutive relations, polarization and magnetization fields. Space-propagating and surface-propagating wave solutions to Maxwell's equations are fundamental to understanding energy and momentum transport around and through nanoscale structured materials. The chapter ends with a development of plane wave propagation in homogenous media and at dielectric and metallic surfaces. This discussion lays the groundwork for the comparison between the waves and transmission lines in Chapter 4.

In addition to the main chapter we have prepared three "complements" which cover some key issues more thoroughly. These complements either extend the exposition to a deeper level or furnish important details and digressions that may be of specific interest to some readers but not to all. By perusing the chapters and complements, the material may be studied at various levels and from various angles. The first complement, Energy Flow in Polarizable Matter, covers the time evolution of energy flux when electromagnetic waves propagate through media with electric polarization. We point out analogies between the behavior of classical fields in bulk matter with the energy dynamics of reactive and dissipative circuits. The second complement, Macroscopic Polarization from Microscopic Polarizability, shows how the macroscopic electric, polarization, and displacement fields can be related to microscopic atomic and molecular properties. The Clausius–Mossotti equation, which expresses the dielectric constant of a material (a macroscopic property) in terms of the microscopic polarizability of the constituent atoms or molecules, is developed at the end of this complement. The Classical Charge Oscillator as Dipole Antenna constitutes Complement 3. In it we show how a "real" antenna can be built up from an array of oscillating charges and how an array of macro-antennas can be used to concentrate the spatial direction of emission or reception.

After these three complements to Chapter 2 we devote Chapter 3 to a fairly extensive discussion of surface waves at the interface between dielectrics and metals, because they play such an important role in "plasmonic" structures and devices. In fact, this propagation can be expressed in terms of circuit and waveguide theory, familiar to electrical engineers; and Chapter 4, Transmission Lines, Waveguides, and Equivalent Circuits, establishes the correspondence between classical electromagnetics and circuit properties such as capacitance, inductance, and impedance. Furthermore we illustrate how waveguide modal analysis and impedance matching can be used to guide the design of nanoscale optical devices.

On the atomic scale (a few hundred picometers) and at interaction energies less than or comparable to the chemical bond, light–matter interaction can be very well understood through a semiclassical approach in which the light field is treated classically and the atom quantally. We therefore retain the *classical* electrodynamics treatment in Chapter 5, Radiation in Classical and Quantal Systems, while presenting a very simple *quantum* atomic structure with dipole transitions among atomic and molecular internal states. We take a very down-to-earth wave mechanical approach to the quantum description in order to bring out the analogies between classical light waves, quantum matter waves, classical dipole radiation and atomic radiative emission.

In addition to the complements we have included a number of appendices that provide some supplementary discussion of the analytical tools used to develop the physics and engineering of light–matter interaction. Appendix A is a brief discussion of systems of units in electricity and magnetism. Although the Système International (SI) has now been almost universally adopted, it is still worthwhile to understand how this system is related to others; which quantities and units can be chosen for "convenience" and which are the universal constraints that all systems must respect. Appendix B is a brief review of vector calculus that readers have probably already seen, but some might find a little refresher discussion useful. Appendix C discusses how the important differential operations of vector calculus can be recast in different coordinate systems. Although the Cartesian system is usually, the most familiar, spherical and cylindrical coordinates are practically indispensable for frequently encountered problems. Much of the book deals with harmonically oscillating fields, and Appendix D is a succinct review of the quite useful phasor representation of these fields. Finally Appendices E, F, and G present the properties of the special functions, Laguerre, Legendre, and Hermite, that are so commonly encountered in electrodynamics and quantum mechanics. These Appendices are an integral part of the book, not just some "boiler plate" nailed on at the end. Readers are strongly encouraged to pay as much attention to them as they do to the chapters and complements.

Most of the material in this book is not new. Excellent texts and treatises on classical electrodynamics, physical optics, circuit theory, waveguide and transmission line engineering, atomic physics, and spectroscopy are readily available. The real aim of this book is take the useful elements from these disciplines and to organize them into a course of study applicable to light–matter interaction at the nanoscale. To the extent, for example, that waveguide mode analysis and sound design practice in microwave propagation informs the nature of light transmission around and through fabricated nanostructures, it is relevant to the purposes of this book. Rugged, reliable laser sources in the optical and near-infrared regime together with modern fabrication technologies at the nanoscale have opened a new area of light–matter interaction to be explored. This exploration is far from complete, but the present book is intended to serve as a point of entry and a useful account of some of the principal features of this new terrain.

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1

Historical Synopsis of Light–Matter Interaction

The phrase "light–matter interaction" covers a vast realm of physical phenomena from classical to quantum electrodynamics, from black holes and neutron stars to mesoscopic plasmonics, nanophotonics, and subatomic quantum objects. The term "interaction" implies that light and matter are distinct entities that influence one another through some intermediate agent. The history of scientific inquiry from the earliest times to the present day might be neatly summarized into three questions: what is the nature of light itself, of matter itself, and of the interaction agent? We now know from Einstein's celebrated equation $E = mc^2$ that light (E) and matter (m) are fundamentally manifestations of the same "thing," related by a universal proportionality constant, the square of the speed of light in vacuum (c^2). Nevertheless under ambient physical conditions normally found on earth, the distinction between light and matter makes sense; their interaction is meaningful and worth studying.

1.1 Light and Matter in Antiquity

In the 5th century BC Leucippus, a Greek philosopher from Miletus (now in Turkey), founded the school of *atomism* in which the universe is composed of immutable, indestructible, indivisible atoms, and the space through which they move, the *void*. His best student was Democritus (460–370 BC) who elaborated the atomistic construct of the universe, attributing natural phenomena to the motion of atoms and the diversity of material objects to their shapes and interlocking structures. The most extensive account of the Leucippus–Democritus atomic theory appears in an extended epic poem, *De rerum natura* (The nature of things) by Lucretius, a Roman who lived much later (99–55 BC). A contemporary of Democritus, the Greek philosopher Empedocles (490–430 BC), proposed that the cosmos was composed of four elements: fire, air, water, and earth. Like the atomist school, these elements were immutable and the diversity of nature arose from their combinations. The dynamics of the combinations are effected by two forces, repulsive and attractive, called strife and love, respectively. Empedocles is also credited with proposing the first theory of light. His idea was that light particles stream out of the eyes and contact material objects. Euclid (~300 BC) assumed that this flux moved in straight lines and used the idea to explain some optical phenomena in Euclid's *Optics*, a very influential early treatise on optics. Euclid's *Optics* in turn influenced Claudius Ptolemy (AD 90–168), a Roman citizen living in Egypt, whose writing on geocentric astronomy was considered definitive until the European Renaissance.

1.2 Light and Matter in the European Renaissance

By the beginning of the 17th century, the certitude of received ideas was crumbling. Earth as the center of the universe and Europe as center of the earth were cast into doubt. The Americas had been discovered by European explorers between 1492 and 1504, the earth had been circumnavigated by 1522, and the Ptolemaic geocentric astronomical system had been effectively overthrown by the Copernican heliocentric revolution of 1543. The invention of the telescope in 1608 enabled Galileo to show that Jupiter's moons revolved around that planet, not the earth. Into this fluid situation stepped René Descartes (1596–1650) with a new world view. Descartes proposed that the universe consisted only of matter and motion. Forces could only be propagated among massive bodies by actual contact, and therefore the apparent space between celestial bodies, the "void" of Democritus, was actually filled with a kind of very-fine-grained material medium or plenum. Light emission, reflection, refraction, and absorption were all explained in terms of material flux. The notion of force "fields" and action at a distance had no place in the Cartesian system of the universe. Descartes's interpretation of refraction, however, was severely challenged by Pierre de Fermat (1601–1665), who explained the deviation of light rays on the basis of the *principle of least time*. Applying this principle, Fermat derived that the sines of the incident and refracted angles are in constant ratio, essentially the equivalent of what we now commonly term "Snell's law." Descartes also derived this law, but his interpretation of light as particle flux required greater velocity in the denser medium whereas the principle of least time imposed a slower velocity. Fermat's principle is in accord with the modern expression for the velocity of light, $v = c/n$ where n, the index of refraction, is unity in free space and greater than unity in material media.

The next significant observation was light "diffraction," a term coined by Grimaldi (1618–1663) to describe the appearance of light beyond the geometrical shadow boundary defined by the supposed rectilinear motion of light-particle flux. Diffraction was also observed by Robert Hooke (1635–1703) who conjectured that light was due to rapid vibratory motion of the very small particles of which ordinary matter is composed. Furthermore, Hooke had the brilliant insight that light (still considered as a kind of matter flux) propagated outward from the center of each tiny vibrating center in circular figures and that light "rays" were trajectories at right angles to these circular figures. This view of light propagation laid the foundation for the construction of wave fronts with which Hooke was able to explain refraction. He also tried to interpret colors in terms of refraction, but his color theory was challenged by Isaac Newton (1642–1727) who correctly interpreted color as an intrinsic property of light and not a distortion of it due to refraction. Although Hooke took the first steps toward a wave theory of light, it was Christiaan Huygens (1629–1695) who put it on a firmer foundation by expressing refraction in terms of the principle that *each element of a wave front may be regarded as the center of a secondary disturbance giving rise to spherical waves. The wave-front at any later time is the envelope of all such secondary wavelets.* Later this principle was refined and extended by the French engineer Augustin-Jean Fresnel (1788–1827) to establish modern wave optics based on the principle of Huygens–Fresnel. It successfully explains light intensity modulations due to diffraction.