Interaction of Electromagnetic Radiation with Matter

（电磁辐射与物质相互作用）

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Foreword

A key discipline situated at the junction of physical optics and condensed matter physics is the interaction of electromagnetic radiation with matter. The variety of different applications of optics in modern physics has led to the understanding that the interaction of electromagnetic radiation with matter has many directions and shades. Firstly, we can mention the interaction of light with matter in metals, semiconductors and insulators. Secondly, the study of optical phenomena such as spontaneous and stimulated absorption and emission of light by matter, the processes of reflection and scattering (i.e., dissipation) of light energy by a substance, in both the classical and the quantum-mechanical approximations, is the main aim of this research of the processes of electromagnetic radiation interaction with matter.

The materials of this book are devoted to the consideration of the first stage of the electromagnetic radiation interaction with different materials: the absorption, scattering, and dispersion of light waves. The description of these phenomena are possible in some cases within the framework of classical electrodynamics. The determination of important optical characteristics of materials, such as the reflection coefficient R, absorption A and the transmittance T, can be carried out on the basis of the macroscopic Maxwell's theory along with Lorentz's microscopic theory.

Let us recall that Maxwell's theory is phenomenological: in this theory, the optical properties of materials are associated with the dielectric permittivity ε, the magnetic permeability μ and the specific conductivity σ of the materials, which are assumed to be known from experience. At the same time, Lorentz's electronic theory determines microscopic electromagnetic fields created by charged particles, which allows us to clarify the physical meaning of the phenomenological constants ε, μ and σ in Maxwell's equations.

Of course, the most complete and consistent description of the processes of electromagnetic radiation interaction with matter can be obtained using the quantum-mechanical description, which allows us to take into account all aspects of the interaction associated with changes in the states of matter and the radiation field. From this point of view, the classical description is a limiting case, which greatly simplifies the consideration of the processes of radiation interaction with matter. The main limitations of applicability of classical descriptions are associated with two circumstances:

• Firstly, the classical representation of radiation in the form of the light waves should be justified.

• Secondly, the classical description ignores the atomic structure of the matter and the dynamics of atomic environment in the substance.

Quantum mechanical theory of the interaction of electromagnetic radiation with matter is indispensable since it gives profound physical insight into the matter especially when high-energy radiation and anisotropy of matter are taken into account and in the case magneto-optical phenomena are included.

This book consists of six chapters. Chapter 1 (Electromagnetic Theory of Light) presents a general introduction to the consideration of Maxwell's system of equations. The solution of these equations allows us to consider a number of important phenomena (e.g., absorption of light by metals), taking into account the limiting circumstances needed for taking the simplified classical approach.

Chapter 2 (Optical Waveguides) states the main physical ideas about the features of electromagnetic radiation propagation in metal and optical waveguides using Maxwell's system of equations as the tool for analysis. Practical applications of optical waveguides for physical data sensors are considered.

Chapter 3 (Optical Processes in the Solid State) gives an overview of some elements of the quantum mechanical theory of the solid state, such as Bloch's theorem, Brillouin's zones, and electron dynamics in the reciprocal lattice.

Chapter 4 (Magneto-optical Phenomena in the Solid State) presents the principles underlying the phenomenological theory of magneto-optical phenomena generalized for all substances, including ferromagnetic metals, magneto-ordered dielectrics and paramagnetic rare-earth (RE) compounds. In addition, the possible mechanisms of the magneto-optical effects arising in the noble and ferromagnetic metals in the semi-classical and quantum mechanical approximations are analyzed.

In Chapter 5 (Magneto-optical Effects in the Rare-earth Compounds), a quantum mechanical theory for the linear magneto-optical effects in paramagnetic rare-earth garnets due to the allowed electric-dipole 4f → 5d transitions is considered in detail. The role of the Van-Vleck "mixing" contribution to the circular magneto-optics of the rare-earth ions in the garnet crystals is discussed. In addition, the basic physical ideas concerning the magneto-optics of emissive 4f → 4f transitions in rare-earth compounds that allows detailed explanation of the spectral features of the magnetic circular polarized luminescence (MCPL) observed for non-Kramers rare-earth ions in the garnet crystals are considered.

In Chapter 6 (Experimental Methods), well-known experimental methods used to determine optical constants of solid states are discussed. The basics of modern modulation spectroscopy and its application for the research of the Van Hove singularities in the optical spectra of metals and semiconductors are considered in detail, and vacuum violet (VUV) spectroscopy using synchrotron radiation sources is included.

Some general questions that arise regarding the theory that is required to explain the data are answered in this book with simple model approximations. Using these simple models makes it easier for readers to understand the physical aspects of the questions that are involved, and at the same time allows the performance of direct mathematical calculations to obtain a final result without complicated mathematical methods. We should like to point out that the text does not represent a comprehensive book on the physics of electromagnetic radiation interaction with matter, which covers a wide range of topics and solid state materials that have not been studied by the present authors.

The book was a joint work of the the authors from three institutes. Chapters 1 to 6 were contributed by Prof. Dejun Fu from the School of Physics and Technology, Wuhan University. Chapter 2 and Chapters 4 through 6 were written by Prof. Uygun V. Valiev from the Department of Photonics, Faculty of Physics, National University of Uzbekistan. Chapters 1, 3, and 5 were written by Prof. Gary W. Burdick from Department of Physics, Andrews University. And Chapters 1, 2, and 4 were written by Dr. Pavel E. Pyak from Department of Theoretical Physics, Faculty of Physics, National University of Uzbekistan.

Contents

Chapter 1

Electromagnetic Theory of Light

1.1 Maxwell's equations

Most optical phenomena occurring in the interaction of electromagnetic radiation with matter and used in optical and laser technologies can be qualitatively and quantitatively explained on the basis of a classical approach. In the classical description, the electromagnetic field is characterized by four main vector quantities: the electrostatic field strength $\boldsymbol{E}$, the electric induction $\boldsymbol{D}$, the magnetic field intensity $\boldsymbol{H}$, and the magnetic induction $\boldsymbol{B}$. Usually the calculation of the electromagnetic field vectors are determined as functions of the spatial coordinates and time. The basic equations describing the propagation of electromagnetic fields in the medium are the macroscopic Maxwell's equations (written in cgs-Gaussian units[1,2])①:

$$\mathrm{div}\boldsymbol{D} = 4\pi\rho \tag{1.1}$$

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① To write the Maxwell's equations in a compact form, the vector-differential operator "nabla" $\boldsymbol{\nabla}$ is used:

$$\boldsymbol{\nabla} = \left[\boldsymbol{i}\frac{\partial}{\partial x} + \boldsymbol{j}\frac{\partial}{\partial y} + \boldsymbol{k}\frac{\partial}{\partial z}\right]$$

Then the operations div and rot applied to some vector $\boldsymbol{A}$ can be represented as:

$$\mathrm{div}\boldsymbol{A} = \boldsymbol{\nabla}\boldsymbol{A} = \frac{\partial A_x}{\partial x} + \frac{\partial A_y}{\partial y} + \frac{\partial A_z}{\partial z}$$

$$\mathrm{rot}\boldsymbol{A} = [\boldsymbol{\nabla}\times\boldsymbol{A}] = \begin{vmatrix} \boldsymbol{i} & \boldsymbol{j} & \boldsymbol{k} \\ \dfrac{\partial}{\partial x} & \dfrac{\partial}{\partial y} & \dfrac{\partial}{\partial z} \\ A_x & A_y & A_z \end{vmatrix} = \boldsymbol{i}\left(\frac{\partial A_z}{\partial y} - \frac{\partial A_y}{\partial z}\right) + \boldsymbol{j}\left(\frac{\partial A_x}{\partial z} - \frac{\partial A_z}{\partial x}\right) + \boldsymbol{k}\left(\frac{\partial A_y}{\partial x} - \frac{\partial A_x}{\partial y}\right)$$

$$\mathrm{div}\boldsymbol{B} = 0 \tag{1.2}$$

$$\mathrm{rot}\boldsymbol{E} = -\frac{1}{c}\frac{\partial \boldsymbol{B}}{\partial t} \tag{1.3}$$

$$\mathrm{rot}\boldsymbol{H} = \frac{4\pi}{c}\left(\boldsymbol{j}_{\mathrm{c}} + \frac{1}{4\pi}\cdot\frac{\partial \boldsymbol{D}}{\partial t}\right) \tag{1.4}$$

The first (1.1) and the second (1.2) Maxwell's equations are a generalization of the Gauss theorem for electric and magnetic fields. The Gauss theorem includes the sources of the fields, with ρ being the charge density. The third Maxwell's equation (1.3) is a generalization of the law of electromagnetic induction: an alternating magnetic field is inseparably linked with the vortex induced electric field. The fourth equation (1.4) is the generalized law of total current, where $\boldsymbol{j}$ is the current density. A magnetic field is generated by a current conduction $\boldsymbol{j}_{\mathrm{c}}$ (i.e., directional movement of electric charges), and equivalently by a displacement current $\boldsymbol{j}_{\mathrm{d}} = \frac{1}{4\pi}\cdot\frac{\partial \boldsymbol{D}}{\partial t}$ (i.e., alternating electrical field).

Maxwell noticed that the system of equations (1.1)–(1.4) not containing the displacement current $\boldsymbol{j}_{\mathrm{d}}$ was not able to describe the presence of the alternating electric (and magnetic) fields. Indeed, applying the divergence operation to equation (1.4) (omitting the displacement current): $\mathrm{div}(\mathrm{rot}\boldsymbol{H}) = 0 = \frac{4\pi}{c}\cdot\mathrm{div}\boldsymbol{j}_{\mathrm{c}}$, then, from the equation $\mathrm{div}\boldsymbol{j} = -\frac{\partial\rho}{\partial t}$ describing the charge conservation law, it follows that the density of electric charges, as well as the values of the electric fields are constant! Therefore Maxwell suggested that the expression $\mathrm{div}\boldsymbol{j} = 0$ is valid for the total current $\boldsymbol{j}$ which is the sum of the conduction current $\boldsymbol{j}_{\mathrm{c}}$ and the displacement current $\boldsymbol{j}_{\mathrm{d}}$, i.e., $\boldsymbol{j} = \boldsymbol{j}_{\mathrm{c}} + \boldsymbol{j}_{\mathrm{d}}$. Then, one can obtain that $\mathrm{div}\boldsymbol{j}_{\mathrm{d}} = -\mathrm{div}\boldsymbol{j}_{\mathrm{c}}$, $\mathrm{div}\boldsymbol{j}_{\mathrm{d}} = \frac{\partial\rho}{\partial t}$. Consequently, in accordance with the Gaussian electrostatic law (1.1), one can obtain that $\mathrm{div}\boldsymbol{j}_{CM} = \frac{1}{4\pi}\cdot\frac{\partial}{\partial t}(\mathrm{div}\boldsymbol{D})$ or $\boldsymbol{j}_{CM} = \frac{1}{4\pi}\cdot\frac{\partial \boldsymbol{D}}{\partial t}$.

The emergence of the displacement current is very important, as it provides the possibility of generating and propagating electromagnetic waves, as it will be shown below in §1.2. Equations (1.1)-(1.4) are not sufficient to describe the electromagnetic field, and they must be supplemented by what are called "material equations". It is assumed that for harmonic electromagnetic fields, which we will consider further, Maxwell's system of equations should be written as follows:

$$\begin{aligned} \boldsymbol{D} &= \varepsilon \boldsymbol{E} = (\boldsymbol{E} + 4\pi \boldsymbol{P}) \\ \boldsymbol{B} &= \mu \boldsymbol{H} \end{aligned} \tag{1.5}$$

where, $\boldsymbol{P} = \alpha \boldsymbol{E}$ is the electrical polarization of the medium, α is the electric susceptibility, μ is the magnetic permeability and ε is the dielectric permittivity. For a linear homogeneous isotropic medium, ε (or μ) is the measure of how easily a material is polarized (or magnetized) in response to an electric (or magnetic) field. The values of the macroscopic theory coefficients ε, σ_1, μ and α depend on the properties of the medium, but we assume they do not depend on the field (the case of a *linear medium*), coordinates (*homogeneous medium*) and direction (*isotropic medium*). However, they may depend on the frequency ω. In addition to equations (1.1)-(1.4) and (1.5), it is necessary to add what is called the "continuity equation" for the current density and Ohm differential law:

$$\mathrm{div}\boldsymbol{j} = -\frac{\partial \rho}{\partial t} \tag{1.6}$$

$$\boldsymbol{j} = \sigma_1 \boldsymbol{E} \tag{1.7}$$

Here σ_1 is the specific conductivity of the medium. Note, that equation (1.6) describes the electric charge conservation law.

To calculate fields in piecewise-continuous mediums such as the air-metal, dielectric-metal, and air-dielectric interfaces, we take into account the boundary conditions, establishing connections between the field vectors from different sides of the interface of the two mediums. The boundary conditions at the interface can be derived from Maxwell's equations written in integral form (see §2.1 below).

1.2 The wave equation in dielectrics, electromagnetic waves and their properties

We can rewrite Maxwell's equations (1.1)-(1.4) for non-conductive $\sigma_1 = 0$ and homogeneous media (i.e., dielectric), in which there are no sources of electric charges (i.e., $\rho = 0$) as follows[2]:

$$\mathrm{div}\boldsymbol{D} = 0 \tag{1.8}$$

$$\mathrm{div}\boldsymbol{B} = 0 \tag{1.9}$$

$$\mathrm{rot}\boldsymbol{E} = -\frac{1}{c}\frac{\partial \boldsymbol{B}}{\partial t} \tag{1.10}$$

$$\mathrm{rot}\boldsymbol{H} = \frac{1}{c}\cdot\frac{\partial \boldsymbol{E}}{\partial t} \tag{1.11}$$

We can apply the operation rot to the third equation (1.10):

$$\mathrm{rot}(\mathrm{rot}\boldsymbol{E}) = -\frac{\mu}{c}\cdot\frac{\partial}{\partial t}(\mathrm{rot}\boldsymbol{H}) \tag{1.12}$$

and replace the right part of the obtained relation by the fourth equation of Maxwell's system (1.11). Then on the one hand, we have

$$\mathrm{rot}(\mathrm{rot}\boldsymbol{E}) = \mathrm{grad}(\mathrm{div}\boldsymbol{E}) - \boldsymbol{\nabla}^2\boldsymbol{E} = -\boldsymbol{\nabla}^2\boldsymbol{E}$$

And on the other hand,

$$-\frac{\mu}{c}\cdot\frac{\partial}{\partial t}(\mathrm{rot}\boldsymbol{H}) = -\frac{\varepsilon\mu}{c^2}\cdot\frac{\partial^2\boldsymbol{E}}{\partial t^2}$$

Equating the right and left parts of the relation (1.12), we obtain the wave equation for the electric vector:

$$\boldsymbol{\nabla}^2\boldsymbol{E} = \frac{\varepsilon\mu}{c^2}\cdot\frac{\partial^2\boldsymbol{E}}{\partial t^2} \tag{1.13}$$

Similarly, one can obtain the wave equation for the magnetic vector:

$$\boldsymbol{\nabla}^2\boldsymbol{H} = \frac{\varepsilon\mu}{c^2}\cdot\frac{\partial^2\boldsymbol{H}}{\partial t^2} \tag{1.14}$$

where $\Delta = \boldsymbol{\nabla}^2 = \dfrac{\partial^2}{\partial x^2} + \dfrac{\partial^2}{\partial y^2} + \dfrac{\partial^2}{\partial z^2}$ is the Laplace operator.

The solutions of equations (1.13) and (1.14) are plane waves:

$$\boldsymbol{E} = \boldsymbol{E}_0\mathrm{e}^{\mathrm{i}(\omega t - \boldsymbol{k}\cdot\boldsymbol{r})} \text{ and } \boldsymbol{H} = \boldsymbol{H}_0\mathrm{e}^{\mathrm{i}(\omega t - \boldsymbol{k}\cdot\boldsymbol{r})} \tag{1.15}$$

where $\boldsymbol{r}$ is the radius vector, ω is the wave frequency, $\boldsymbol{k}$ is the wave vector (or wave number) which is equal to $|\boldsymbol{k}| = \dfrac{2\pi}{\lambda}$, where λ is the wavelength in the medium. These waves propagate in the medium with velocity $v = \dfrac{c}{\sqrt{\varepsilon\mu}}$, where c is the velocity of electromagnetic wave in vacuum (approximately, $c = 3 \times 10^{10}\text{cm/s}$). Then the refractive index n of a medium for electromagnetic waves is equal to $n = \sqrt{\varepsilon\mu}$.

Substituting the plane wave vectors $\boldsymbol{E}$ and $\boldsymbol{H}$ from (1.15) into the material equations (1.5) for the vectors $\boldsymbol{D}$ and $\boldsymbol{B}$, one can obtain the transverse condition for electromagnetic waves: $\boldsymbol{k}\cdot\boldsymbol{E} = 0$ and $\boldsymbol{k}\cdot\boldsymbol{H} = 0$. That is, longitudinal components of the electric and magnetic fields are absent, meaning the fields are directed perpendicular to the direction of propagation of the electromagnetic waves. In addition, in an electromagnetic wave, the vectors $\boldsymbol{E}$ and $\boldsymbol{H}$ are orthogonal to each other ($\boldsymbol{E}\perp\boldsymbol{H}$) and satisfy the proportionality relationship $\sqrt{\varepsilon} \cdot E = \sqrt{\mu} \cdot H$. The orthogonality condition of vectors $\boldsymbol{E}$ and $\boldsymbol{H}$ in an electromagnetic wave can be obtained as follows: substitute equation (1.15) into the third equation of the system of Maxwell's equations (1.10), where the wave vector of the wave can be written as: $\boldsymbol{k} = k \cdot \boldsymbol{n}$, where $\boldsymbol{n}$ is a unit vector and $k = \dfrac{\omega}{c}\sqrt{\varepsilon(\omega)} = k_0\sqrt{\varepsilon(\omega)}$. The relationship between the vectors $\boldsymbol{E}$ and $\boldsymbol{H}$ can be found taking into account the additional condition $\mu = 1$[3], from third equation of system of equations (1.3):

$$\text{rot}\boldsymbol{E} = -\mathrm{i}[\boldsymbol{k} \times \boldsymbol{E}] = -\mathrm{i}k[\boldsymbol{n} \times \boldsymbol{E}] = -\mathrm{i}\frac{\omega\sqrt{\varepsilon}}{c}[\boldsymbol{n} \times \boldsymbol{E}] \tag{1.16}$$

$$-\frac{1}{c} \cdot \frac{\partial \boldsymbol{H}}{\partial t} = -\mathrm{i}\frac{\omega}{c}\boldsymbol{H}$$

Equating the left and right sides of the original equation, one can obtain $\sqrt{\varepsilon}\cdot[\boldsymbol{n}\times\boldsymbol{E}] = \boldsymbol{H}$. Therefore, the vector obtained from the vector product of the perpendicular vectors $\boldsymbol{n}$ and $\boldsymbol{E}$ is oriented perpendicular to both of them, and therefore, one can see that $\boldsymbol{E}\perp\boldsymbol{H}$.

It is well known that for finding the values of grad, div and rot for non-stationary vector fields-electric and magnetic-defined for each point in space, it is convenient to introduce some auxiliary quantities, such as the scalar potential φ and the vector potential $\boldsymbol{A}$ of the electromagnetic field. We know that the gradient of the scalar potential φ determines the electrostatic field strength, and the absence of magnetic charges means that the magnetic vector $\boldsymbol{H}$ can be defined as the rot of a vector $\boldsymbol{A}$ (since $\mathrm{div}\boldsymbol{H} = \mathrm{div}(\mathrm{rot}\boldsymbol{A}) = 0$), which is called the vector-potential of the electromagnetic field. The time derivative of this vector will determine the variable (vortex) electric field strength. Taking into account that the permeability μ in the optical region of the spectrum is approximately equal to unity[3], we find that:

$$\mathrm{rot}\boldsymbol{E} = -\frac{1}{c}\cdot\frac{\partial \boldsymbol{H}}{\partial t} = -\frac{1}{c}\cdot\frac{\partial}{\partial t}(\mathrm{rot}\boldsymbol{A}) = -\frac{1}{c}\cdot\mathrm{rot}\left(\frac{\partial \boldsymbol{A}}{\partial t}\right), \quad \text{i.e., } \boldsymbol{E} = -\frac{1}{c}\left(\frac{\partial \boldsymbol{A}}{\partial t}\right) \tag{1.17}$$

In other words, in contrast to electrostatics, the vector of a non-stationary electric field having a vortex character cannot be represented in the form of the gradient of a scalar potential. Rather, it can be expressed via a superposition of the scalar and the vector potentials according to the formula:

$$\boldsymbol{E} = -\mathrm{grad}\varphi - \frac{1}{c}\left(\frac{\partial \boldsymbol{A}}{\partial t}\right) \tag{1.18}$$

It is interesting to note that the introduction of the electromagnetic field vector-potential $\boldsymbol{A}$ allows one to define a generalized momentum $\boldsymbol{P}$ of a particle with charge e in an electromagnetic field as follows:

$$\boldsymbol{P} = \boldsymbol{p} + \frac{e}{c}\boldsymbol{A} \tag{1.19}$$

where $\boldsymbol{p}$ is the mechanical momentum, and the second term is due to the interaction of the charged particle with the vortex electric component of the electromagnetic field. Indeed, let's consider the force acting on a charge in an electric field $\boldsymbol{E}$:

$$\boldsymbol{F} = e\boldsymbol{E} = -e\cdot\mathrm{grad}\varphi - \frac{e}{c}\left(\frac{\partial \boldsymbol{A}}{\partial t}\right)$$

It can be seen that this force consists of two parts. The first is due to the action of the stationary part of the electric field, which causes the change in the mechanical momentum $\boldsymbol{p}$, and the second is due to the action of the vortex part of the electric field, which contributes to the mechanical momentum $\boldsymbol{p}$:

$$\boldsymbol{F} = \frac{\mathrm{d}\boldsymbol{p}}{\mathrm{d}t} - \frac{e}{c}\left(\frac{\partial \boldsymbol{A}}{\partial t}\right) = \frac{\mathrm{d}}{\mathrm{d}t}\left(\boldsymbol{p} + \frac{e}{c}\boldsymbol{A}\right)$$

In other words, when moving in an electromagnetic field, the particle has a generalized momentum $\boldsymbol{P}$, rather than the mechanical momentum $\boldsymbol{p}$.

1.3 Conducting medium and Hagen-Rubens relation

Before proceeding to a discussion of the wave equation for a conducting homogeneous medium (i.e., for $\boldsymbol{j} \neq 0$), it is necessary to clarify the question about the existence of electric charges in a conducting medium at high frequencies.

For this, let us apply the operation of div to the fourth equation of the system of Maxwell's equations (1.1)-(1.4)[2]:

$$\mathrm{div}(\mathrm{rot}\boldsymbol{H}) = 0 = \frac{4\pi}{c}\mathrm{div}\boldsymbol{j} + \frac{1}{c}\cdot\frac{\partial}{\partial t}(\mathrm{div}\boldsymbol{D})$$

and using (1.6), one can obtain

$$\left(\frac{4\pi\sigma_1}{c}\right)\cdot\left(\frac{4\pi}{\varepsilon}\right)\cdot\rho + \left(\frac{4\pi}{c}\right)\cdot\frac{\partial\rho}{\partial t} = 0$$

Thus, we have a first order differential equation:

$$\frac{\partial\rho}{\partial t} + \left(\frac{4\pi\sigma_1}{\varepsilon}\right)\rho = 0 \tag{1.20}$$

Using separation of variables: $\dfrac{\mathrm{d}\rho}{\rho} = -\left(\dfrac{4\pi\sigma_1}{\varepsilon}\right)\mathrm{d}t$, its solution can be found as:

$$\rho(t) = \rho_0\cdot\mathrm{e}^{-\left(\frac{4\pi\sigma_1}{\varepsilon}\right)t} \tag{1.21}$$

where $\rho_0 = \rho(t=0)$.

Using literature values of electrical quantities for copper, $\varepsilon = 10$, $\sigma_1 = 5\times 10^{17}\mathrm{s}^{-1}$[4], and taking into account that the time interval is on the order of magnitude $t \approx T = 10^{-15}\mathrm{s}$, we see that the quantity in the exponent in relation (1.21) is on the order of 10^2!

In other words, the value of ρ in a conducting medium at high frequencies is negligible, and assuming that in the optical region of the spectrum $\mu \approx 1$[3], the