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1

Introduction

The way in which light interacts with material objects is determined by the optical properties of the materials. Why might you want to think about these optical properties? There are at least two reasons. First, you can make use of known optical materials to design and build devices to manipulate light: mirrors, lenses, filters, polarizers, and a host of other gadgets. Second, you can measure the optical properties of some new material and obtain a wealth of information about the low energy excitations that govern the material's physics. Figure 1.1 is a chart that identifies some of these excitations and indicates the part of the spectrum where they might be expected to appear.

In common parlance, "optical" can be a synonym of "visual," and hence related to human eyesight. This interpretation would restrict discussion to the visible part of the electromagnetic spectrum, light with wavelengths of 390–780 nm, indicated by the little rainbow in Fig. 1.1. (I will need to introduce the variety of measures used for the wavelength or the frequency or the photon energy of electromagnetic waves. Four are shown in Fig. 1.1: λ in μ m, *f* in THz, *E* in meV, and another frequency unit, wavenumbers or cm⁻¹. The latter may be unfamiliar to persons who have not worked in the field of optical effects in solids; it is the inverse of the wavelength in cm. The visible spectrum spans 385–770 THz, 1.59–3.18 eV, and 12,800–25,600 cm⁻¹. Units used in papers about the optical properties of solids are discussed in Appendix A.)

Of course I will *not* in this book make the interpretation that optical means visible; instead, materials properties will be considered over a wide range of frequencies or wavelengths.* Plausible ranges are discussed toward the end of this chapter.

Even if you were constrained to the use of your own eyes, you would see that solids have a wide range of optical properties. Silver is a lustrous metal used for centuries in coins and fine tableware, with a high reflectance over the whole visible range. Silicon is a crystalline semiconductor and the foundation of modern electronics. With its surface oxide freshly etched off, silicon is also rather reflective, although not as good a mirror as silver.[†] Salt (sodium chloride) is a transparent ionic insulator, is necessary for life, and makes up about 3.5% (by weight) of seawater. A crystal of salt is transparent over the entire visible spectrum; because the refractive index is about 1.5, the reflectance is everywhere about 4%.

If you had ultraviolet eyes, you would see these materials differently. Silver would be a poor reflector, with at most 20% reflectance and trailing off to zero at the shortest wavelengths. In contrast, the reflectance of silicon would be better than in the visible,

1

^{*} I like the notion of "DC to daylight," used widely in the amateur radio community and also as the name of a symposium honoring Professor A.J. Sievers, at Cornell University, June 14, 2003.

[†] Silver reflects about 98% of red light and about 80% of violet light; silicon reflects about 33% of red and 50% of violet.

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Fig. 1.1

Chart showing optical processes in solids, with an indication of the wavelengths or frequencies where these processes typically may be studied. The uppermost scale shows the vacuum wavelength in μ m, 10⁻⁶ m. Frequencies (or energies) are given on three scales. Top to bottom they are: THz, 10¹² cycles/s; photon energies, meV; and wavenumbers, $\tilde{\nu}$ (in cm⁻¹), defined by $\tilde{\nu} = 1/\lambda$ with λ the wavelength measured in cm.

reaching up to 75%. Sodium chloride would be opaque over much of the spectrum, with a reflectance a bit higher than in the visible. Those with infrared eyes would also see things differently from visible or UV-sensitive individuals. Silver would have a reflectance above 99%. Silicon would appear opaque at the shortest infrared wavelengths but would then become transparent, so that you could see through even meter-thick crystals.* Sodium chloride remains transparent over much of the infrared, but an opaque and highly reflecting "reststrahlen" (German for residual ray) region occurs at long wavelengths. In the reststrahlen band, NaCl has a reflectivity not much below that of a metal.

If you put on your solid-state-physics hat, you can understand the optical properties of these materials, at least qualitatively. Silver is a nearly free-electron metal, with one electron per atom in the metallic Fermi surface. These mobile electrons give the high electrical conductivity; they form a plasma that makes silver opaque and highly reflective

^{*} Here ultra-high-purity is assumed. Moreover, in the middle infrared region there is a band caused by lattice vibrational effects – multiphonons in this case – where silicon is opaque unless rather thin.

3

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Introduction

below the plasma frequency.* Silicon is a semiconductor with a gap between the filled valence band and the empty conduction band. Photons with energies below the gap can propagate without loss in silicon. Photons with energies above the gap are absorbed, generating electron–hole pairs. This absorption renders silicon opaque and, as mentioned, increases the reflectance. Sodium chloride is an insulating crystal, with a band gap in the ultraviolet. Similar to silicon, photons with energy larger than the gap are absorbed. Sodium chloride has two atoms per unit cell; these occur as ions, Na^+ and Cl^- ; an electric field displaces these ions, producing induced dipoles in the solid. With a two-atom basis, the lattice vibrations have an optical branch, and the reststrahlen band is a result of the light exciting this optical branch.

Now let me return to the question of the range of wavelengths (or the range of light frequencies or of photon energies) over which I can discuss the optical properties of solids. The electromagnetic spectrum extends over a huge range; a representative cartoon illustrating the "electromagnetic spectrum" is shown in Fig. 1.2.

This chart shows wavelengths from km to pm along with corresponding frequencies and photon energies. So the question is: What part of this spectrum might be used to study the optics of solids?

To start, I'll want to use continuum electrodynamics, so the short wavelength limit is set by a requirement that the wavelength be larger than the spacing between atoms. When the wavelength is less than the interatomic distances, diffraction effects dominate. X-ray diffraction is essential for determining crystal structure but beyond my scope. At somewhat longer wavelengths, continuum electrodynamics is fine, but the materials properties are essentially a superposition of atomic transitions. Solid-state effects contribute of course but minimally for wavelengths shorter than something on the order of 50 nm.[†]

As wavelengths get longer and longer, there is of course no problem with continuum electrodynamics. However, practically speaking, the physics that govern the electromagnetic response at dc and audio frequencies is the same as the physics at ultra-high radio frequencies and even microwaves. So the lowest frequencies that I will consider are around a few GHz.

There is a second reason for setting a long-wavelength limit. One GHz (4 μ eV photon energy) corresponds to 30 cm wavelength, and this large scale raises an experimental issue: to shine light on a solid, one sends a beam that one would like to consider to be composed of plane electromagnetic waves. Cartoons of typical experimental setups are shown in Fig. 1.3. The left panel shows a reflectance (\Re) experiment and the right a transmittance (\mathscr{T}) experiment. Light comes from a source (of known properties) that can emit a range of wavelengths, encounters the sample, and goes to a detector where it is converted to

^{*} The connection between high absorption, high conductivity, and high reflectance is not intuitive. For the moment, I'll just assert that all three go together. So at wavelengths where a material is opaque, it also has increased reflectance. The more intense is the absorption, the higher the conductivity and also the higher the reflectance. A hand-waving argument says that high conductivity means large currents in response to applied electric fields; the power loss or absorption goes as $\mathbf{j} \cdot \mathbf{E} = \sigma E^2$. See Section 4.5 for further discussion.

[†] Using $\lambda f = c$, $\tilde{\nu} = 1/\lambda$, and E = hf with λ the wavelength, f the frequency in Hz, $\tilde{\nu}$ the frequency in cm⁻¹, or wavenumber, E the photon energy, c the speed of light, and h Planck's constant, 50 nm corresponds to a frequency of 6 PHz (petaHertz), a wavenumber of 200,000 cm⁻¹, and photon energies of 25 eV.

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wavelength of the frequency. These ideas are only a good approximation to experiment when the wavelength is small compared to the size of the sample or of the experimental apparatus. When this condition is no longer the case, diffraction effects (by the sample not by the atomic lattice) as well as waveguide effects in the surrounding apparatus become important.*

* One may of course measure materials properties all the way to zero frequency (infinite wavelength) by electrical means: apply contacts and measure resistance, capacitance, etc. I will discuss the connection of optical measurements to dc electrical properties a number of times.

5

Cambridge University Press 978-1-107-16014-9 — Optical Effects in Solids David B. Tanner Excerpt <u>More Information</u>

Introduction

It makes little sense to be very precise in specifying the wavelength or frequency limits over which optical concepts are important for the physics of solids. So I'll say that I will consider the wavelength range to be several cm to several tens of nm, the frequency range to be a few GHz to a few PHz, and the photon energy range to be tens of μ eV to tens of eV. This range covers the bands labeled microwaves, far infrared, midinfrared, near infrared, visible, and ultraviolet in Fig. 1.2. There is a factor of a million between one end and the other; that should be enough for everybody.*

In the following chapters, I'll remind you a little bit about electromagnetism, including Maxwell's equations and their plane-wave solutions. I'll then restrict myself for some time to local, nonmagnetic, isotropic, homogeneous, and linear solids.[†] The motivations for these restrictions are that the materials should be local, so that the current at point \mathbf{r} is a function only of the fields at \mathbf{r} ; nonmagnetic, because most solids are nonmagnetic and because even magnetic materials only show themselves to be magnetic at rather low frequencies; isotropic, so that the properties do not depend on the direction or polarization of the light; homogeneous, so that the response functions do not depend on spatial position; and linear, so that I may make a Fourier decomposition of the fields and treat each component independently. With these approximations, I'll introduce the idea of a complex dielectric function, discuss classical theories of free carrier response in metals, interband absorption in semiconductors and insulators, and lattice vibrations (phonons). Next, I will show data from the literature to illustrate some of the concepts of these theories.

I return to electromagnetism to calculate the reflection and transmission by a thin film or slab as a way to link experimental measurements to the optical properties of the material making up the film. The next step is to introduce simple quantum mechanics, leading to a discussion of free-electron metals, followed by a presentation of the quantum-mechanical perturbation-theory of optical absorption, culminating in an important sum rule for the conductivity. The sum rule result motivates an interlude about causality where I obtain the Kramers–Kronig relations between the absorptive and dispersive parts of the response function, discuss the analysis of reflectivity by Kramers–Kronig methods, and derive other sum rules. My focus shifts back to materials, with a simple treatment of the optics of superconductivity, a distinctly quantum-mechanical phenomenon. Next comes the band structure of simple solids and the interband absorption edge in semiconductors, followed by materials with strong correlations and interactions.

After this, it will be time to relax the initial conditions and discuss (one at a time!) nonlocal properties, mostly the anomalous skin effect in a pure metal, wave propagation in anisotropic materials, magneto-optics, and randomly inhomogeneous materials.[‡] Several appendices discuss units, some mathematics, and other things "optical."

^{*} In fact there are few materials that have been studied over the entire range. A much more typical range is in wavelength from, say, 0.3 mm to 300 nm, far infrared to ultraviolet, a range of 10^3 .

[†] Much of this discussion can apply to liquids as well, and even to a dilute gas, but the physics discussion will rely on solid-state physics ideas: Fermi surfaces, band structure, etc.

[‡] I'll leave the huge subjects of quantum optics and nonlinear optics to others; I think it is better to say nothing than to make short and probably superficial treatments.

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2

Maxwell's Equations and Plane Waves in Matter

2.1 Optical Constants

The response of materials to light is described by a number of quantities, often called "optical constants." Among these are:

- ϵ , the dielectric constant.
- σ , the electrical conductivity.
- X, the susceptibility.*
- *n*, the refractive index.
- κ , the extinction coefficient.
- δ , the electromagnetic skin depth.
- *Z*, the surface impedance.

and many others. See Appendix F for a longer but still incomplete list.

These quantities are neither constant nor independent. They are functions of the frequency, temperature, pressure, external magnetic field, and many other things. By knowing two of these, one that describes the absorption in the solid (such as the electrical conductivity or the extinction coefficient) and one that describes dispersion (such as the dielectric constant or the refractive index), all of the others can be calculated.

2.2 Maxwell's Equations

In my initial Electricity and Magnetism course, the professor said that the subject is governed by equations written in the nineteenth century by Maxwell [1] and that some day a teacher would come in, write these equations on the blackboard on the first day of the class and then proceed to develop a theory based on these equations. Of course in a junior-level class, he did not do this, but I can do it here.

There are two versions: Maxwell's microscopic equations and Maxwell's macroscopic equations. The first are more fundamental, because they describe the microscopic fields arising from every charge in the Universe and from the motion of these charges as well.

* Electric and magnetic.

6

7

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2.2 Maxwell's Equations

The second are more fun, because they average over the charges in macroscopic media* and allow great simplification of the subject.

After restricting myself to macroscopic charges and currents, I have ρ_{ext} , the external charge density, and \mathbf{j}_{free} , the free current density as sources in Maxwell's equations. The external charge density is essentially the charge imbalance in the medium; it is zero for electrically neutral objects.[†] The free current density is the result of the motion of free charges in the metal. The standard example of these mobile charges is the free electrons in a metal, but could also be doped or thermally excited free carriers in a semiconductor or the diffusion of ions in an electrolyte. Note that the free carriers are compensated by bound ions in the electrically neutral material so these charges do not generate any external charge density.

So let me begin by writing Maxwell's equations for macroscopic media [2–8]. I'll use cgs-Gaussian units; the translation to SI units appears in Appendix B.

 ∇

$$\nabla \cdot \mathbf{D} = 4\pi \rho_{\text{ext}} \tag{2.1a}$$

$$\mathbf{f} \cdot \mathbf{B} = 0 \tag{2.1b}$$

$$\nabla \times \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t}$$
(2.1c)

$$\nabla \times \mathbf{H} = \frac{4\pi}{c} \mathbf{j}_{\text{free}} + \frac{1}{c} \frac{\partial \mathbf{D}}{\partial t},$$
(2.1d)

I must add a connection to classical mechanics to these equations. The force \mathbf{F} on a particle with electric charge q satisfies the Lorentz force law,

$$\mathbf{F} = q \left(\mathbf{E} + \frac{\mathbf{v}}{c} \times \mathbf{B} \right), \tag{2.2}$$

where \mathbf{v} is the particle's velocity vector.

The definitions of the auxiliary fields are

$$\mathbf{D} = \mathbf{E} + 4\pi \mathbf{P}$$
$$\mathbf{H} = \mathbf{B} - 4\pi \mathbf{M}.$$
 (2.3)

The quantities in Eqs. 2.1, 2.2, and 2.3 are all functions of space and time. The vector **E** is called the "electric field" and the vector **D** is called the "electric displacement field." In vacuum, **D** and **E** are proportional to each other, with the multiplicative constant ϵ_0 depending on the physical units.[‡] Inside a material they are different on account of the polarization of the material. The vector **P** is the "polarization field," also known as the "electric polarization," "electric polarization density," or "electric dipole moment/unit volume." The vector **B** is called the "magnetic field," and it and **E** are defined as the vector

^{*} In the absence of external fields, the positive charges (the nuclei) in a uniform medium are perfectly screened by the negative charges (the electrons) so that both may be omitted completely from the charges and currents in Maxwell's equations. External fields may polarize these charges (pushing + in one direction and - in the opposite direction); the effects of such currents and dipole moments are the subject of our optical properties studies.

[†] And it does *not* include any charge imbalance caused by external fields. This polarization gets included in the dipole moment/unit volume **P**.

[‡] Unity in cgs-Gaussian units.

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8

Maxwell's Equations and Plane Waves in Matter

fields necessary to make the Lorentz law correctly describe the forces on a moving charged particle. **B** is also called the "magnetic flux density," or the "magnetic induction." The vector **H** is also sometimes called the "magnetic field." Other names include the "magnetic field intensity," the "magnetic field strength," and the "magnetizing field." In vacuum, **B** and **H** are proportional to each other, with the multiplicative constant μ_0 depending on the physical units.* Inside a material they are different on account of the magnetization of the material. The scalar ρ_{ext} is the "free volume charge density" and the vector **j** free is the "free electric current density." The vector **M** is the "magnetization field" or the "magnetic dipole moment/unit volume."

Three of the four Maxwell equations have names: Equation 2.1a is Gauss' Law; Eq. 2.1c is Faraday's law of induction; and Eq. 2.1d is Ampère's law with Maxwell's correction. I'll just call it Ampère's law. Eq. 2.1b is sometimes called Gauss' law for magnetism but is sometimes called the no-magnetic-monopole law. Others say that it has no name. I like "the no-monopole law."

2.3 Total, Free, and Bound Charges and Currents

If I view charges as being small point-like particles moving here and there in space, then a classical-physics-style definition of the microscopic charge density in a solid might be

$$\rho_{\text{micro}}(\mathbf{r},t) = \sum_{i} q_i \delta(\mathbf{r} - \mathbf{r}_i(t))$$

whereas the current would be written

$$\mathbf{j}_{\text{micro}}(\mathbf{r},t) = \sum_{i} q_{i} \mathbf{v}_{i}(t) \delta(\mathbf{r} - \mathbf{r}_{i}(t)).$$

Here, the sum *i* runs over all charged particles, q_i is the charge of the *i*th particle, located at (time-dependent) coordinate \mathbf{r}_i and moving with (time-dependent) velocity \mathbf{v}_i .

There are quantum mechanical versions of the charge density and current density also. To use them requires a solution of Schrödinger's equation in the solid. With the solution in hand, I could then find the contribution of the electrons[†] (with charge -e) to the total charge density to be $\rho_e = -e\Psi_e^*\Psi_e$ where Ψ_e is some total wave function of the electrons, and, similarly $\mathbf{j}_e = -(\hbar e/2mi) [\Psi_e^* \nabla \Psi_e - (\nabla \Psi_e^*)\Psi_e]$.

Accounting for all the particles in a material with 10^{22} particles per cubic centimeter is of course too hard, so I'll average over some volume ΔV . The scale of V is taken to be large with respect to the interatomic spacing a and small with respect to the electromagnetic wavelength λ , or

$$a^3 \ll \Delta V \ll \lambda^3$$
.

^{*} Unity in cgs-Gaussian units.

[†] Because of a choice made by Franklin, the electron is negative. I'll take *e* to be a positive number and put the sign in explicitly as needed.

9

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2.4 Maxwell's Equations for Solids

The inequalities are easy to satisfy because $a \sim 0.1$ nm and, according to the discussion in Chapter 1, λ is bigger than 10 nm, so there is a factor of 10⁶ between their cubes. The charge and current then are written as averaged quantities, $\rho(\mathbf{r},t) = qn(\mathbf{r},t)$ and $\mathbf{j}(\mathbf{r},t) = \rho(\mathbf{r},t)\mathbf{v}_d(\mathbf{r},t)$, where q is the average charge of the particles, n is their number density, and \mathbf{v}_d the average velocity in the volume ΔV , known as the "drift velocity."

The electrons and nuclei do not know that I would like to average them out, so they still respond to applied fields, producing dipole moments in the material, **P** and **M**. These bound or polarization charges, electric polarization currents, and magnetization currents are determined by the physics of the solid. Then, the macroscopic polarization charge density ρ_{pol} , the polarization current density \mathbf{j}_{pol} , and the magnetization current density \mathbf{j}_{mag} are defined in terms of polarization **P** and magnetization **M** as $\rho_{pol} = -\nabla \cdot \mathbf{P}$, $\mathbf{j}_{pol} = \partial \mathbf{P}/\partial t$, and $\mathbf{j}_{mag} = c\nabla \times \mathbf{M}$.

Maxwell's macroscopic equations reduce to the microscopic equations if one recognizes that the microscopic, free, and bound charge and current density are related by $\rho_{\text{micro}} = \rho_{\text{ext}} + \rho_{\text{pol}}$, and $\mathbf{j}_{\text{micro}} = \mathbf{j}_{\text{free}} + \mathbf{j}_{\text{pol}} + \mathbf{j}_{\text{mag}}$, and then uses Eq. 2.3 to eliminate **D** and **H**.

2.4 Maxwell's Equations for Solids

I am almost to a point where I can begin to address optical effects in solids. But there are two things still to do. First, the world is in general electrically neutral,* so I will take $\rho_{\text{ext}} = 0$. For notational convenience, I will drop the "free" subscript on the current, with the understanding that it is the free current that is meant.[†] Thus, $\mathbf{j}_{\text{free}} \rightarrow \mathbf{j}$.

Henceforth, I'll use the form of Maxwell's macroscopic equations written just below. I'll claim that these cover the most general cases that occur in the studies of optical effects in solids. I'll make further simplifications in the next several pages, but can always return to these as a starting point.

$$\nabla \cdot \mathbf{D} = 0 \tag{2.4a}$$

$$\nabla \cdot \mathbf{B} = 0 \tag{2.4b}$$

$$\nabla \times \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t}$$
(2.4c)

$$\nabla \times \mathbf{H} = \frac{4\pi}{c} \mathbf{j} + \frac{1}{c} \frac{\partial \mathbf{D}}{\partial t}$$
(2.4d)

The four parts of Eq. 2.4 have a certain pleasing symmetry. After I've defined and used the complex dielectric function, this symmetry will become more perfect.

^{*} Anyone who has been shocked by a static discharge after crossing a rug on a dry day or who has seen a lightning strike from a thunderstorm knows (1) that this statement is not always true but (2) that the charge imbalance does not last.

[†] And see the discussion in Section 3.1 where the distinction between free and bound currents will be blurred as well.

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10

Maxwell's Equations and Plane Waves in Matter

2.5 Plane-Wave Solutions

I write the electric field as a complex exponential

$$\mathbf{E}(\mathbf{r},t) = \mathbf{E}_0 e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)}$$
(2.5)

and also the magnetic field

$$\mathbf{H}(\mathbf{r},t) = \mathbf{H}_0 e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)},\tag{2.6}$$

where \mathbf{E}_0 and \mathbf{H}_0 are constant vectors giving the amplitudes of the fields (complex quantities in the most general case) and their directions,* \mathbf{q} is the wave vector of the field (measured in cm⁻¹ in cgs), and ω is the angular frequency (in radians/s or s⁻¹). I choose to use **H** rather than **B**, as does Jackson [2].

The quantity $i = \sqrt{-1}$ does not appear in Eqs. 2.1–2.4. Hence the only mechanism to have equations that contain real and imaginary quantities is through writing the fields as complex quantities, as in Eqs. 2.5 and 2.6. It would be perfectly valid to write $\mathbf{E} = \mathbf{E}_0 \cos(\mathbf{q} \cdot \mathbf{r} - \omega t + \phi_e)$ and $\mathbf{H} = \mathbf{H}_0 \cos(\mathbf{q} \cdot \mathbf{r} - \omega t + \phi_m)$, where the phases ϕ_e and ϕ_m allow for differing phases in electric and magnetic fields.[†] In this case, all quantities in electromagnetism would be purely real quantities. I could do this; at a minimum it would give fine training in the use of trigonometric identities, because as soon as I take a derivative, there will be both sines and cosines in the math.

It is conventional to say that one writes the fields as complex quantities but when one wants to evaluate the observable fields, the real part should be taken. This statement is basically true, though one has to be careful in cases where two complex fields are multiplied together.

I know from freshman physics that the crests and valleys of the wave repeat in space (at fixed time) by a translation of the wavelength λ . From this assertion I get $|\mathbf{q}|\lambda = 2\pi$ or $|\mathbf{q}| = 2\pi/\lambda$. At a point in space, the wave repeats every time the time advances by the period T; hence $\omega = 2\pi/T = 2\pi f$ with f the frequency. Moreover, $\lambda f = v$ and $\omega = qv$ with v the wave speed (= c in vacuum). See Appendix A for a discussion of units used for length, time, frequency, energy, fields, and many other quantities used in optical studies.

The use of plane-wave fields may seem arbitrary or restrictive, but Fourier tells us that

$$\mathbf{E}(\mathbf{r},t) = \int d^3q \int d\omega \, \mathbf{E}_0(\mathbf{q},\omega) e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)},$$

with

$$\mathbf{E}_0(\mathbf{q},\omega) = \int d^3r \int dt \, \mathbf{E}(\mathbf{r},t) e^{-i(\mathbf{q}\cdot\mathbf{r}-\omega t)}.$$

I can write an arbitrary field in terms of the Fourier integral, do the usual trick of exchanging order of integration and differentiation, and find (for linear, local materials) that Maxwell's equations apply to each Fourier component.

- * Hence, to be explicit I could write $\mathbf{E}_0 = \hat{e}\mathcal{E}_0 e^{i\phi_0}$, where \hat{e} is a unit vector pointing in the field direction, \mathcal{E}_0 is the field magnitude, and ϕ_0 is a constant phase which, when combined with $(\mathbf{q} \cdot \mathbf{r} \omega t)$, specifies where the zeros, crests, and valleys of the wave occur.
- [†] Alternatively, write $\mathbf{E} = \mathbf{E}_c \cos(\mathbf{q} \cdot \mathbf{r} \omega t) + \mathbf{E}_s \sin(\mathbf{q} \cdot \mathbf{r} \omega t)$ and a similar equation for **H**.