in the classic theory is equally not a gauge-invariant quantity. However, the mean value of the velocity of a particle,

\[ \mathbf{\bar{v}}_i = \frac{1}{m_i} \left( -i \hbar \mathbf{\nabla}_i - \frac{c_i}{c} \mathbf{A} - \frac{\mathbf{\ell}_i}{c} \mathbf{A}(\mathbf{e}) \right) \]

is gauge-independent; this can be confirmed by straightforward calculation. The same is true for any power of velocity and, hence, for any operator which is expandable in a series in powers of \( r \) and \( \mathbf{r} \).

1.3. Averaging of Maxwell's Equations

The true values of the fields \( \mathbf{e} \) and \( \mathbf{h} \) in a medium vary in a very complicated manner in space and time owing to the uninterrupted motion of all particles; this motion generates rapidly fluctuating local densities of charges and currents. Moreover, if the description is at a microscopic level, where quantum effects are essential, it is not possible at all to assign to each particle definite values of coordinates and velocities; one can only operate with the probability for particles to have specific coordinates and velocities. Strictly speaking, it is simply meaningless to try to determine the instantaneous local values of the fields \( \mathbf{e} \) and \( \mathbf{h} \). Any sensible theory can operate only with the values, averaged over fluctuations, of fields and with their correlations at different spacetime points, or with the probability distribution for various field configurations.

The Maxwell–Lorentz equations (1.1.1) are linear and thus allow direct averaging, which reduces to simple replacement of \( \mathbf{e} \) and \( \mathbf{h} \) by their mean values \( \mathbf{E} \) and \( \mathbf{H} \). The charge and current densities are then also replaced with the mean values. Obviously, the relations between currents and fields (the so-called material equations) can be nonlinear; however, this is a different problem which has no direct bearing on the problem of averaging of the field equations in the form (1.1.1).

The averaged Maxwell's equations are written in the form

\[
\begin{align*}
\text{curl} \mathbf{H} &= \frac{1}{c} \frac{\partial \mathbf{E}}{\partial t} + \frac{4\pi}{c} \mathbf{j} \\
\text{curl} \mathbf{E} &= -\frac{1}{c} \frac{\partial \mathbf{H}}{\partial t} \\
\nabla \mathbf{E} &= 4\pi \mathbf{\rho} \\
\nabla \mathbf{H} &= 0
\end{align*}
\] (1.3.1)

The same notation as in nonaveraged equations (1.1.1) is used above for the averaged values of the current density \( \mathbf{j} \) and charge \( \mathbf{\rho} \).

Owing to the linearity of equations (1.1.1), they can be averaged by physically very different methods. Thus the procedure of averaging the Maxwell–Lorentz equations over the so-called physically infinitely small volume has been known for about a century now, since Lorentz's time. It is assumed in this procedure that the concept of field at each point must be discarded, and that the long-wavelength field (with \( \lambda \gg a_0 \)) can be regarded as uniform within volumes whose linear size is much less than \( \lambda \).

If the chosen, physically infinitely small volume contains a large number of particles, the volume can be treated as macroscopic. The statistical fluctuations of the field averaged over a volume are small and one can regard the field averaged over a physically infinitely small volume as the statistically mean field. In this way, one can construct the macroscopic electrodynamics and optics.

This approach is not satisfactory for our purposes, because

1. The wavelength in the medium may be considerably reduced (owing to a high refractive index); moreover, there exist qualitatively new phenomena, such as gyrotropy, which are defined only by the drop of the field over molecular-scale distances,

2. The condition \( \lambda \gg a_0 \) is not satisfied at all in the x-ray range, but the most important reason is that

3. To find the motion of particles in a medium and their response to fields, one needs to know not the average field but the true field at points where particles are.

The averaging over a physically infinitely small volume immediately excludes from analysis the field acting on a particle and forces one to forego the response of the medium to the field; after this, one has to be satisfied with taking only a phenomenological account of this response.

Therefore, averaging over a physically infinitely small volume has to be dropped from a microscopic theory of matter’s response to electromagnetic field and one has to resort to the method, standard for the statistical physics, of averaging over the ensemble of the possible states of the medium, for example, over the Gibbs distribution. In the quantum case, this approach implies also the averaging over wave functions of particles. In fact, charge and current densities are thereby averaged, and the corresponding mean fields are automatically found from equations (1.3.1). This averaging is carried out over fluctuations, not over volume. By virtue of the ergodic hypothesis, this also eliminates temporal fluctuations since the statistical averaging is equivalent to averaging over time.

This does not mean that the information on fluctuations is lost completely. We can still write equations for correlation functions of the type \( \varepsilon_\alpha(r,t)\varepsilon_\beta(r',t') \) when the mean fields are zero, as in the case of thermal radiation. Again, some averaged characteristics of the fluctuation ensemble are taken into account.

It is important that there is no spatial averaging and the fields are
referred to a point (to within a length of the order of $10^{-13}$, as we have mentioned above).

It is thus sufficient, owing to the linearity of Maxwell's equations, to average their right-hand sides that contain $j$ and $\rho$; note that the charge and current densities are averaged by averaging over the Gibbs ensemble. If external fields are also present and $j^{(e)}$ and $\rho^{(e)}$ are their sources, then $j(r, t)$ and $\rho(r, t)$ represent the medium's response to these external fields. In their turn, these also induce fields in the medium, so that the next problem is to establish the relation of the induced currents $j$ and charges $\rho$ to the external fields. As the next step, averaged equations

\[
\begin{align*}
\text{curl} \mathcal{H} &= \frac{1}{c} \frac{\partial \mathcal{E}}{\partial t} + \frac{4\pi}{c} j + \frac{4\pi}{c} j^{(e)} \\
\text{curl} \mathcal{E} &= \frac{1}{c} \frac{\partial \mathcal{H}}{\partial t} \\
\nabla \mathcal{E} &= 4\pi \rho + 4\pi \rho^{(e)} \\
\nabla \mathcal{H} &= 0
\end{align*}
\]

must be used to find the relation between the currents $j$ and the mean fields in the medium,

\[
j = j(\{E\})
\]

The relation between the currents $j$ and the field $\mathcal{E}$ in the medium dictates such characteristics of the medium as its linear and nonlinear susceptibilities.

The refusal to average fields over physically infinitely small volumes results in a significant restructuring of the entire approach to describing the electromagnetic field in matter. The standard procedure of macroscopic electrodynamics is to single out the physically different components in the current $j$,

\[
j(r, t) = j_f + j_b + j_m
\]

where $j_f$ is the current of free charges (conduction electrons, etc.), $j_b = \partial P / \partial t$ is the polarization current, or the displacement current (by definition, $P$ is the medium's polarization vector) due to the motion of bound charges, and $j_m = c \text{curl} \mathcal{M}$ is the magnetization vortex current (by definition, $\mathcal{M}$ is the magnetization vector). The mean value of the vector $h$, denoted above as $\mathcal{H}$, is usually referred to as the magnetic induction $B$; also introduced are the electric induction vector

\[
P = \mathcal{E} + 4\pi \mathcal{P}
\]

and the magnetic field strength vector

\[
\mathcal{H} = B - 4\pi \mathcal{M}
\]

As a result, we obtain ordinary equations of macroscopic electrodynamics for the vectors $\mathcal{E}$, $\mathcal{H}$, $\mathcal{D}$, and $\mathcal{B}$, which must be appended with two phenomenological relations between these vectors.

This procedure is justified in the range of quasistationary currents but is not valid for the optical and still shorter wavelength range, where unambiguous decomposition of current into $j_f$, $j_b$, and $j_m$ is not possible and thus becomes meaningless.

Indeed, we can only distinguish between free and bound charges, or distinguish between closed (vortex) and open currents, if they are considered in a volume that is greater than the region of localization of the bound charges or of closed current loops. At each individual point, however, these contributions to current are essentially identical to one another: they are all contained in the common expression (1.1.2).

We shall illustrate this general statement with several simple examples. At low frequencies, the separation of charges into free and bound types is obvious: free charges are displaced by external fields to macroscopic distances, while bound charges stay within their respective atoms or molecules, and their small displacements within these bounds produce only a local polarization. However, this difference vanishes at high frequencies. Indeed, the classical equation of motion of free charges in a field $E$ at a frequency $\omega$,

\[
m \ddot{\mathbf{r}} = eE \cos \omega t
\]

gives the oscillation amplitude of the order of $eE/m\omega^2$. Even in the strong field of a ruby laser ($\omega \approx 3 \times 10^{10} \text{ s}^{-1}$), with the electric field strength of the order of $E \sim 10^5 \text{ CGS} = 3 \times 10^7 \text{ V cm}^{-1}$, the oscillation amplitude is of the order of $10^{-8} \text{ cm}$, and is even lower in the weak field of ordinary light.

A bound electron is characterized by a very similar amplitude of motion (of the order of one Bohr radius). Consequently, the currents of free and bound electrons cannot be separated.

Another example is the photoelectric effect, in which bound electrons are transferred to the free state as a result of absorption of a quantum of light. At high frequencies, light quanta knock out bound electrons from atoms and thus convert them from bound into free particles. The question is this: to which current should we assign the contribution of these transition processes, that of the free or that of the bound charges? This example again demonstrates the arbitrariness of separating the current into distinct parts.

We find the same situation with the magnetization current, which typically involves the motion of charges along closed trajectories. At low frequencies, this motion is driven by the magnetic field but if electrons are subjected to circularly polarized light, they move along circular trajectories of radius $eE/m\omega^2$, which, as shown above, can be of the order of $a_0$. Free charges in electric field then contribute to the magnetization current just
as bound electrons do. It is thus clear that it is impossible and physically meaningless to strictly separate the magnetization current and the current of free charges (even though one can always identify the total vortex component of the current; it is not determined by magnetic field alone).

The only way left to us in the case of high-frequency fields is to operate with the total current \( j \) which describes all processes. The equations for the mean fields \( \mathcal{E} \) and \( \mathcal{H} \) then take the form (1.3.1) or (1.3.2). These equations are written identically to those used in the electrodynamics of metals but \( j \) and \( \rho \) are now not the current and charge densities of free particles but the total current and the corresponding charges.

Sometimes the total polarization \( \mathcal{P}(r,t) \) is used instead of the total current \( j(r,t) \); formally, it is defined as

\[
\frac{\partial \mathcal{P}}{\partial t} \quad \mathcal{P}(r,t) = \int_\infty^t j(r,t')dt'
\]

that is, it contains the contributions of both free and bound charges and of magnetization currents. The lower bound of integration must be taken as \( t = -\infty \) (we assume that neither fields nor polarization existed in the distant past).

Now we can also introduce the total induction

\[
\mathcal{D}(r,t) = \mathcal{E}(r,t) + 4\pi \mathcal{P}(r,t)
\]

just as this is done for bound charges.

The equations can now be rewritten as

\[
\begin{align*}
\text{curl} \mathcal{H} &= \frac{1}{c} \frac{\partial \mathcal{D}}{\partial t} + \frac{4\pi}{c} j^{(e)} \\
\text{curl} \mathcal{E} &= -\frac{1}{c} \frac{\partial \mathcal{H}}{\partial t} \\
\nabla \mathcal{D} &= 4\pi \rho^{(e)} \\
\nabla \mathcal{H} &= 0
\end{align*}
\]

(1.3.3)

The third equation is obtained using the continuity equation

\[
\frac{\partial \rho}{\partial t} = -\nabla j = -\nabla \frac{\partial \mathcal{P}}{\partial t}
\]

whence

\[
\rho = -\nabla \mathcal{P}
\]

as we have it in electrostatics.

---

**General Theory of Interaction of Electromagnetic Fields with Matter**

Equations (1.3.3) are similar to the ordinary set of equations of the optics of dielectrics; however, with the definition chosen above, the induction \( \mathcal{D} \) includes both the effects of motion of free charges and the magnetization currents. Thus there is no need to introduce magnetic induction. The vector \( \mathcal{P} \) implies magnetic phenomena as well (this will be shown later in the text).

Both approaches (equations (1.3.2) and (1.3.3)) are completely equivalent. We can therefore use either of the approaches: to construct the theory of "metallic" type using matter equations of the type

\[
\mathbf{j} = \sigma \mathcal{E}
\]

or the theory of "dielectric" type, using the relation between the total polarization and the field,

\[
\mathcal{P} = \chi \mathcal{E}
\]

Since \( \mathcal{D} \) includes all the currents, only one susceptibility \( \chi \) is introduced; there is no need in introducing magnetic susceptibility.

---

**1.4. Expansion of Current in Powers of Field**

Our main objective now is to calculate the current induced in a system by an external field. In the general case, the problem of calculating the mean current in an arbitrary field and for an arbitrary system of a macroscopic number of particles has no solution.

Only one regular approach is known which allows an analysis of the solution in the general form. It involves the expansion of the response in powers of field:

\[
\mathbf{j}(r,t) = \sum_{k=0}^{\infty} j^{(k)}(r,t) \quad j^{(k)}(r,t) \propto \mathcal{E}^k
\]

The ordinary electrodynamics is linear:

\[
\mathbf{j} = \sigma \mathcal{E} \quad \mathcal{P} = \chi \mathcal{E}
\]

Formerly, optics was also limited to linear terms. In the most general terms, the linear relation between coordinates- and time-dependent current vectors and the field has the form

\[
\mathbf{j}_a^{(1)}(r,t) = \int \mathbf{a}(r',t',t') \mathcal{E}_b(r',t')d^3r'dt'
\]

(1.4.1)

This formula is a linear relation between the current and the electric field \( \mathcal{E} \). The magnetic field is not involved here since it can be expressed
in terms of \( \mathcal{E} \) using the second Maxwell’s equation in (1.3.1) or (1.3.2). Furthermore, its direct effect on charges is smaller by a factor \( v/c \) than that of the electric field.

With the advent of lasers, many nonlinear effects were discovered in optics. They can be described only by taking into account higher-order terms in the expansion of current in powers of field. In the general case, these nonlinear terms look as follows:

\[
J^{(2)}(r, t) = \int \sigma^{(2)}_{\alpha \beta \gamma}(r, r', t', t, t') \mathcal{E}_\beta(r', t') \mathcal{E}_\gamma(t'', t'') \, d^3r' \, d^3r'' \, dt' \, dt''
\]

\[
J^{(3)}(r, t) = \int \sigma^{(3)}_{\alpha \beta \gamma \delta}(r, r', r'', t', t'', t', t'') \mathcal{E}_\beta(r', t') \times \mathcal{E}_\gamma(t'', t'') \mathcal{E}_\delta(r'', t'') \, d^3r' \, d^3r'' \, dt' \, dt'' \, dt'''
\]

Terms of fourth and higher orders in field have similar form.

We can make use of the expansion in powers of field as long as the external fields are much lower than the internal Coulomb fields, so that perturbations cannot greatly change the system and its properties:

\[
\mathcal{E} / \mathcal{E}_{at} \ll 1
\]

Here \( \mathcal{E}_{at} \sim e / a_0^2 \sim 10^9 \text{ V cm}^{-1} \) is a typical interatomic field. A field of the order of \( \mathcal{E}_{at} \) destroys atoms and molecules over times of the order of atomic time scale, that is, \( \sim 10^{-16} \text{ s} \). Fields of this strength are hardly ever produced in laboratories; matter can only exist in such fields as a plasma, with a different nonlinearity parameter. The nonlinearity parameter \( \mathcal{E} / \mathcal{E}_{at} \) can be used for gases, liquids, and nonconducting solids. In the case of plasmas, metals, and semiconductors, on the other hand, all of which contain free charges, the possibility of expanding the current in a series in powers of field requires additional analysis. We will discuss it later. If the expansion of current in a series in powers of field is found not to be valid, for example, for the resonance interaction, one resorts to various simplified models which make it possible to solve the system of equations exactly, without power expansion.

By analogy to (1.4.1)–(1.4.3), we can write a series expansion of polarization in powers of field:

\[
P_\alpha(r, t) = \int \chi^{(1)}_{\alpha \beta}(r, r'; t', t') \mathcal{E}_\beta(r', t') \, d^3r' \, dt' + \int \chi^{(2)}_{\alpha \beta \gamma}(r, r', r'', t', t', t'') \mathcal{E}_\beta(r', t') \mathcal{E}_\gamma(r'', t'') \, d^3r' \, d^3r'' \, dt' \, dt'' + \ldots
\]

It is necessary to comment on the limits of integration in (1.4.1)–(1.4.4). By virtue of the causality principle, the current and polarization at a time \( t \) depend only on the values of fields at earlier moments of time. Consequently, integration in time — in \( t', t'' \), etc. — is made from \( -\infty \) to \( t \). Owing to the constraints due to relativity, the integration in space is carried over the region in which \( |r - r'| \leq c|t - t'| \) since the values of fields at points which do not satisfy this condition cannot influence the current at a point \( r \) at a time moment \( t \) (no interaction can propagate at a speed above the speed of light). As a rule, however, the finiteness of the limits of integration over \( r' \) and \( r'' \), etc. is unimportant. It is acceptable to integrate in \( r' \) with infinite limits because a substantially smaller volume of space really contributes to the integral: the response functions \( \sigma \) and \( \chi \) fall off to almost zero over distances \(|r - r'| \) that are much smaller than the indicated relativistic limits.

As we see from expressions (1.4.1)–(1.4.4), the current or polarization at a point \( r \) at a time moment \( t \) are determined in the general case by the values of the field \( \mathcal{E} \) at other spatial points \( r' \) — one then speaks of the nonlocality of the response, and by the values at preceding moments of time \( t' \) — this is known as retardation.

The nature of the retardation effect is sufficiently obvious. A particle “remembers” for some time the field at the preceding moments of time. For example, a free charge remembers the acceleration, and hence the field, of the preceding moments during the time \( \tau \) of velocity relaxation. For a bound electron, this time interval is of the order of the atomic time scale

\[
T_{at} \sim \hbar / m \sim 10^{-16} \text{ s}
\]

that is, of the time during which the velocity of an atomic electron remains unchanged. This time is of the order of the period of optical radiation waves, so that retardation is significant in the optical and still shorter wavelength ranges. Figure 1.1 shows a typical dependence of \( \sigma^{(1)} \) or \( \chi^{(1)} \) on \( t - t' \).

Nonlocality is a less familiar phenomenon. It arises because a particle arrives at a point \( r \) from \( r' \) and carries the memory of the action exerted on it at the point \( r' \). Consequently, the nonlocality radius for free charges, that is, the distance \(|r - r'| \) on which the response functions \( \sigma \) and \( \chi \) fall off almost to zero, is found to be of the order of the free path length \( l = v \tau \). For bound charges, on the other hand, it is of the order of atomic or molecular size \( a_0 \) (Figure 1.2). The significance of the nonlocality effect depends on the ratio of the nonlocality radius to the characteristic distance over which the field changes significantly. Typically, phenomenological electrodynamics of continuous media uses as this distance the wavelength \( \lambda \).

For the optical and the ultraviolet wavelength ranges, and all the more so for the infrared and radio ranges, we have \( \lambda > a_0 \sim 10^{-8} \text{ cm} \). Consequently, if \(|r - r'| \leq a_0 \), the field at a point \( r' \) at such frequencies is almost
identical to the field at a point \( r \). This is the reason why the nonlocality effect is usually only secondary while retardation is almost always significant. There are cases, however, when nonlocality is decisive (long free path lengths in metals or plasma, polaritons in semiconductors and dielectrics).

The nonlocality radius for a free charge is determined by the smaller of two lengths: the free path length \( l = \nu \tau \) or the displacement of a charge over one field oscillation period \( \nu \lambda/c \). If the nonlocality period is dictated by the second of the two lengths, the ratio of the nonlocality radius to the wavelength is of the order of \( \nu/c \ll 1 \). Nevertheless, exceptional cases are possible here even if the speed of light \( c/n \) is low (near its absorption band of the matter) and comparable with that of a particle \( \nu \); nonlocality is then strong.

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Sometimes nonlocality must be taken into account even if it is small. Thus gyrotropy (the effect of rotation of the plane of polarization in matter) is a corollary of nonlocality. In the framework of the approach we have chosen, it will be shown that magnetic susceptibilities are manifestations of nonlocality.

All nonlocality effects vanish after averaging over a physically infinitely small volume of a size much larger than \( a_0 \) or \( v \tau \). This is an additional argument in favor of rejecting this type of averaging.

As for the microscopic approach to the description of the electromagnetic field in a medium, the nonlocality effects are always significant. It will become clear later, especially when crystals are considered, that the propagation of even long-wavelength field in condensed media is always accompanied with a small-scale field structure whose amplitude is not low and which originates from the fields of individual particles; these last fields are induced by the main field. The wavelengths characteristic of this structure are precisely of the order of interparticle distance, \( a_0 \), so that nonlocality effects in it are definitely not small. This structure determines the familiar phenomenon: the field acting on each particle in the medium differs from the mean field, and this must always be taken into account in a microscopic analysis of response.

1.5. Linear Susceptibilities and Their Properties

In the general case, the material equations (1.4.1)–(1.4.4) for stationary and spatially uniform media are integral and can be transformed to algebraic form using the Fourier transform.

A medium is said to be stationary if its properties are invariant in time in the absence of strong time-dependent fields. The choice of the origin \( t = 0 \) of the frame of reference is arbitrary; time enters the response of the medium only as the difference \( t - t' \),

\[
\sigma_{\alpha\beta}^{(1)}(r, r'; t - t') \quad \chi_{\alpha\beta}^{(1)}(r, r'; t - t')
\]

By definition, the spatial uniformity implies that the properties of the medium are identical at all its points. If this is so, \( \sigma_{\alpha\beta}^{(1)}(r, r'; t - t') \) is not altered by the displacement \( r \rightarrow r + \rho, r' \rightarrow r' + \rho \):

\[
\sigma_{\alpha\beta}^{(1)}(r, r'; t - t') = \sigma_{\alpha\beta}^{(1)}(r + \rho, r' + \rho; t - t')
\]

This requirement is equivalent to stating that \( \sigma_{\alpha\beta}^{(1)} \) is a function only of the difference in coordinates:

\[
\sigma_{\alpha\beta}^{(1)}(r, r'; t - t') = \sigma_{\alpha\beta}^{(1)}(r - r'; t - t')
\]
The spatial uniformity is a property characterizing liquids, gases and amorphous solids, but not crystals that we will discuss separately. If the field and all other quantities are expanded in Fourier integrals,

$$E_\alpha(r, t) = \int E_{k\omega} e^{i k \cdot r - \omega t} d^3 k d\omega/(2\pi)^4$$  \hspace{1cm} (1.5.1)$$

and inversely,

$$E_{k\omega} = \int E_\alpha(r, t) e^{-i k \cdot r + \omega t} d^3 r dt$$

we obtain

$$j_{k\omega}^{(1)} = \sigma_{\alpha\beta}^{(1)}(k, \omega) E_{k\omega}$$ \hspace{1cm} (1.5.3)$$

where $$\sigma_{\alpha\beta}^{(1)}(k, \omega)$$ is the Fourier transform of the function $$\sigma_{\alpha\beta}^{(1)}(r - r', t - t')$$:

$$\sigma_{\alpha\beta}^{(1)}(k, \omega) = \int_{0}^{\infty} \int \sigma_{\alpha\beta}^{(1)}(R, \tau) e^{-i k \cdot R + \omega \tau} d\tau d^3 R$$

and $$R = r - r';$$ also, $$\tau = t - t' > 0,$$ which reflects the causality principle: the action must precede the response.

We have mentioned already that instead of the current density, it is often more convenient to operate with polarization $$P,$$ and instead of electric conduction $$\sigma^{(1)},$$ the susceptibility $$\chi^{(1)}.$$ Since

$$j(r, t) = \frac{\partial P(r, t)}{\partial t}$$

or, in terms of Fourier transforms,

$$j_{k\omega} = -i \omega P_{k\omega}$$

then

$$P_{k\omega}^{(1)} = \chi_{\alpha\beta}^{(1)}(k, \omega) E_{k\omega}$$ \hspace{1cm} (1.5.4)$$

where

$$\chi_{\alpha\beta}^{(1)}(k, \omega) = \frac{i}{\omega} \sigma_{\alpha\beta}^{(1)}(k, \omega)$$  \hspace{1cm} (1.5.5)$$

Relation (1.5.5) between $$\chi$$ and $$\sigma$$ is not limited to the case of linearity because it has been obtained from the general relation between current and polarization.

Now we can also consider the dielectric permittivity

$$\varepsilon_{\alpha\beta}(k, \omega) = \delta_{\alpha\beta} + 4\pi \chi_{\alpha\beta}^{(1)}(k, \omega)$$

which relates induction $$D$$ to the electric field strength $$E$$ when we use Maxwell’s equation (1.3.3) written in the “dielectric” form.

For nonuniform media, we can consider functions of the type $$\chi_{\alpha\beta}^{(1)}(r, \omega),$$ which is the Fourier transform of $$\chi_{\alpha\beta}^{(1)}(r', \tau)$$ in time only. One refers to the dependence of $$\sigma, \chi$$ and $$\varepsilon$$ on frequency as their dispersion, and to their dependence on $$k,$$ as to their spatial dispersion.

The dispersion is a corollary of the retardation effect, with the spatial dispersion following from nonlocality.

In optics, and in electrodynamics in general, the frequency of radiation is $$\omega = \omega n,$$ where $$n$$ is the refractive index of a medium; it may seem at the first glance, therefore, that the number of independent parameters of the Fourier transform decreases. The actual situation is, however, that $$\omega$$ and $$k$$ play here the role of formal parameters of the Fourier transform and are thus independent variables. Relations of the type (1.5.3) must be essentially valid not only for a free field which is a superposition of plane waves with $$\omega = \omega n$$ but for any fields (e.g., static fields) produced by an arbitrary distribution of charges and currents.

Fields, currents, and polarizations are real, which imposes constraints on the Fourier transforms. Since $$E(r, t)$$ and $$j(r, t)$$ are real, definitions (1.5.2) and (1.5.4) imply that

$$E_{k\omega} = E_{-k, -\omega, \alpha}^{*} \quad j_{k\omega} = j_{-k, -\omega, \alpha}^{*} \quad P_{k\omega} = P_{-k, -\omega, \alpha}^{*}$$

Hence,

$$\sigma_{\alpha\beta}^{(1)}(k, \omega) = \sigma_{\alpha\beta}^{(1)}(-k, -\omega)$$ \hspace{1cm} (1.5.6)$$

$$\chi_{\alpha\beta}^{(1)}(k, \omega) = \chi_{\alpha\beta}^{(1)}(-k, -\omega)$$

When we later derive the general microscopic expression for susceptibility and conduction, we will be able to prove a less trivial proposition

$$\chi_{\alpha\beta}^{(1)}(k, \omega) = \chi_{\alpha\beta}^{(1)}(-k, -\omega)$$ \hspace{1cm} (1.5.7)$$

which is equivalent to the symmetry principle for kinetic coefficients.

In any specific medium, other symmetry properties are found which impose additional constraints on the tensors $$\chi$$ and $$\sigma.$$ In the general case of stationary and spatially uniform medium, however, it is impossible to derive any other symmetry properties except those given above, unless one looks into the specific structure of the medium.

Now we can check whether anything has been lost (magnetic effects, etc.) when, in contrast to the traditional approach, only the induction is introduced. To do this, we look at the low-frequency range where conduction currents, displacement currents, and magnetization currents are clearly separated, that is, we make use of the quasistationary currents approximation.
First we write the expression for the total current neglecting magnetization currents:

\[ j_\alpha(t) = \tilde{\sigma}_{\alpha\beta} E_\beta + \frac{\partial}{\partial t} (\tilde{\chi}_{\alpha\beta} E_\beta) = \frac{\partial \mathcal{P}_\alpha}{\partial t} \]

We also neglect here both retardation and nonlocality, since the wavelength is in the low-frequency range \( \lambda \gg l \), where \( l \) is the sample size; \( \tilde{\sigma}_{\alpha\beta} \) is the conductivity at low frequencies due to free charges, and \( \tilde{\chi}_{\alpha\beta} \) is the dielectric susceptibility due to bound charges. The tensors \( \tilde{\sigma} \) and \( \tilde{\chi} \) are real.

For the total polarization we obtain

\[ \mathcal{P}_\alpha(t) = \tilde{\chi}_{\alpha\beta} E_\beta(t) + \int_{-\infty}^{t} \tilde{\sigma}_{\alpha\beta} E_\beta(t') \, dt' \]

and in terms of the Fourier transforms,

\[ P_{\omega\alpha} = \tilde{\chi}_{\alpha\beta} E_{\omega\beta} + \frac{i}{\omega} \tilde{\sigma}_{\alpha\beta} E_{\omega\beta} = \left( \tilde{\chi}_{\alpha\beta} + \frac{i}{\omega} \tilde{\sigma}_{\alpha\beta} \right) E_{\omega\beta} \]

Comparing it with the general relation in the approach chosen,

\[ P_{\omega\alpha} = \chi^{(1)}_{\alpha\beta}(\omega) E_{\omega\beta} \]

we obtain

\[ \chi^{(1)}_{\alpha\beta}(\omega) = \tilde{\chi}_{\alpha\beta} + \frac{i}{\omega} \tilde{\sigma}_{\alpha\beta} \quad (1.5.8) \]

In what follows, it is shown that \( \tilde{\chi} \) corresponds to the non-dissipative component of the total susceptibility \( \chi \), and \( i\tilde{\sigma}/\omega \) corresponds to the dissipative component.

Therefore, the contribution of the free and bound charges is not lost in the approach described. Let us look now at how magnetic currents are described when only induction and only susceptibility are used. It may seem at the first glance that the magnetization currents are lost since only the electric field is found in expressions for current, such as (1.4.1). In actual fact, however, the electric and magnetic fields are related through Maxwell’s equations.

Let us take standard expressions for the magnetization current,

\[ j_m(t) = c \text{curl} \mathcal{M} = c \text{curl} \tilde{x}_m (\mathcal{H} - 4\pi \mathcal{M}) \approx c \text{curl} \tilde{x}_m \mathcal{H} \]

For the sake of simplicity, we will consider the case of isotropic medium, which is characterized by scalar magnetic susceptibility \( \tilde{x}_m \) at low frequencies.

Let us make use of Maxwell’s equation

\[ \text{curl} \mathcal{E} = -\frac{1}{c} \frac{\partial \mathcal{H}}{\partial t} \]

Integrating it in time and assuming that all fields are zero at \( t = -\infty \), we obtain

\[ \mathcal{H}(r, t) = -c \int_{-\infty}^{t} \text{curl} \mathcal{E}(r, t') \, dt' \]

Therefore, the magnetization current can be written in the form

\[ j_m(r, t) = -c^2 \tilde{x}_m \int_{-\infty}^{t} \text{curl} \text{curl} \mathcal{E}(r, t') \, dt' \]

\[ = -c^2 \tilde{x}_m \int_{-\infty}^{t} \left( \text{grad} \nabla \mathcal{E} - \Delta \mathcal{E} \right) \, dt' \]

In Cartesian coordinates, this vector relation is rewritten as

\[ j_{m\alpha}(r, t) = -c^2 \tilde{x}_m \int_{-\infty}^{t} \left( \frac{\partial^2}{\partial x_\alpha \partial x_\beta} - \frac{\partial^2}{\partial x_\gamma \partial x_\eta} \delta_{\alpha\beta} \right) \mathcal{E}_\beta dt' \quad (1.5.9) \]

We will show now that a contribution of the form of (1.5.9) can be singled out from the general expression (1.4.1). To do this, we separate from the total current its component which contains second derivatives of electric field with respect to spatial coordinates. This can be done only if the nonlocality effect, that is, spatial dispersion, is taken into account.

Let us assume that nonlocality effects are small, that is, that the characteristic length of nonuniformity of the field (this is typically the wavelength) is much greater than nonlocality radius (as a rule, it is of the order of \( a_0 \)). In this case, the response \( a_{\alpha\beta}^{(1)}(r - r', t - t') \) vanishes only where \( |r - r'| \lesssim a_0 \).

Let us expand \( \mathcal{E}_\beta(r', t') \) in the integrand of formula (1.4.1) into a Taylor series in the neighborhood of a point \( r \):

\[ \mathcal{E}_\beta(r', t') = \mathcal{E}_\beta(r, t) + \frac{\partial \mathcal{E}_\beta(r, t)}{\partial x_\gamma} (x'_\gamma - x_\gamma) + \frac{1}{2} \frac{\partial^2 \mathcal{E}_\beta(r, t)}{\partial x_\gamma \partial x_\delta} (x'_\gamma - x_\gamma)(x'_\delta - x_\delta) + \ldots \]
If this expansion is substituted into (1.4.1), the field \( \mathbf{E}(r, t') \) and its derivatives can be factored out of the integral in spatial variables. Let us introduce the following notation:

\[
\begin{align*}
\sigma^{(1)}_{\alpha\beta}(t - t') &= \int \sigma^{(1)}_{\alpha\beta}(r - r', t - t') \, d^3 r' \\
a_{\alpha\beta\gamma}(t - t') &= \int (x'_{1} - x_{1})(x'_{2} - x_{2}) \sigma^{(1)}_{\alpha\beta\gamma}(r - r', t - t') \, d^3 r' \\
b_{\alpha\beta\gamma\delta}(t - t') &= \int (x'_{1} - x_{1})(x'_{2} - x_{2}) \sigma^{(1)}_{\alpha\beta\gamma\delta}(r - r', t - t') \, d^3 r'
\end{align*}
\]

This gives

\[
\begin{align*}
\mathbf{j}_{\alpha}(r, t) &= \int_{-\infty}^{t} \left\{ \sigma^{(1)}_{\alpha\beta}(t - t') \mathbf{E}_{\beta}(r, t') + a_{\alpha\beta\gamma}(t - t') \frac{\partial \mathbf{E}_{\beta}(r, t')}{\partial x_{\gamma}} \\
&\quad + \frac{1}{2} b_{\alpha\beta\gamma\delta}(t - t') \frac{\partial^2 \mathbf{E}_{\beta}(r, t')}{\partial x_{\gamma} \partial x_{\beta}} + \ldots \right\} \, dt'
\end{align*}
\tag{1.10}
\]

The first term in braces is the response of the system to the field if the spatial dispersion is neglected. We have already discussed it.

For media that are symmetric with respect to inversion, the second term containing \( \partial \mathbf{E}_{\beta}/\partial c_{\gamma} \) vanishes since \( a_{\alpha\beta\gamma} = 0 \).

Note that the third term has a contribution corresponding to magnetic currents. If we assume

\[
b_{\alpha\beta\gamma\delta} = -2\chi_{m}c^2(k_{\alpha\gamma}k_{\beta\delta} - \delta_{\alpha\beta}\delta_{\gamma\delta})
\]

we thereby single out from (1.10) a contribution which coincides with the expression for the magnetic current, (1.9).

In the approach chosen here, the magnetization current is a corollary of weak spatial dispersion. It is the induction current due to the second derivatives of electric field with respect to coordinates. The ratio of the magnitude of this current to the main contribution (the first term in (1.10)) is of the order of \( (a_{0}/\lambda)^2 \sim (v_{sw}/c)^2 \ll 1 \). This estimate explains why the magnetic susceptibility (for paramagnetic materials, \( \chi_{m}^{(1)} \sim 10^{-9} \)) is small in comparison with the susceptibility of dielectrics (\( \chi_{0} \sim 1 \)). We were thus able to show that expressions of the type of (1.4.1) include all currents: conduction, displacement and magnetization ones.

Let us switch to the Fourier representation in the expression (1.5.9) for the magnetization current. We obtain

\[
j_{m\omega\alpha} = -2\chi_{m}c^2 \frac{i}{\omega} \left( \delta_{\alpha\beta} - \frac{k_{\alpha}k_{\beta}}{k^2} \right) \mathbf{E}_{\omega \beta} \tag{1.11}
\]

The expression

\[
\Pi_{\omega\alpha\beta}(\delta_{\alpha\beta} - k_{\alpha}k_{\beta}/k^2)
\]

is the operation of projecting on the directions perpendicular to \( k \); it singles out only the transverse fields. This corresponds to the fact that the magnetization current is determined only by the transverse (vortex) component of the electric field. Formula (1.5.11) implies an expression for the contribution to \( \chi^{(1)}_{m}(k, \omega) \) due to magnetization currents. Taking into account (1.5.3) and (1.5.6), we derive from (1.5.11) that

\[
\chi^{(1)}_{m}(k, \omega) = \frac{\varepsilon_{m}^{2}k_{m}^{2}\chi_{m}}{\omega^2} \left( \delta_{\alpha\beta} - \frac{k_{\alpha}k_{\beta}}{k^2} \right) \tag{1.12}
\]

Note that this contribution to the susceptibility \( \chi^{(1)} \) at low frequencies has a singularity \( \omega^{-2} \). The origin of this singularity is purely formal. The corresponding vortex electric field \( \mathbf{E} \sim \partial \mathbf{H}/\partial t \) is very low and is proportional to the frequency so that the formula for susceptibility includes the factor \( \omega^{-1} \). The second factor \( \omega^{-1} \) originates in definition (1.5).

### 1.6. Relation of Dissipation of Energy in a Medium to the Anti-Hermitian Component of Linear Susceptibility

We have seen already that the imaginary component of susceptibility (1.5.8) is determined at low frequencies in the range of quasistationary currents by the static electric conductivity of the medium, that is, is related to the dissipation of the field's energy. We can now show that this meaning of the imaginary component \( \chi \) is retained at all frequencies.

The dissipation of the energy of the field is the work that the field does over the particles of the medium. The energy dissipated per unit time in unit volume of a region \( V \) whose dimensions are much larger than the nonlocality radius is

\[
Q = \frac{1}{V} \int_{V} \mathbf{j}_{\alpha}(r, t) \mathbf{E}_{\alpha}(r, t) \, d^3 r \tag{1.6.1}
\]

It is not easy to speak of losses at a point when nonlocality is taken into account; hence, integration over the volume \( V \) is carried out. The dissipation, as well as the response of the medium, becomes locally meaningless. For harmonic fields and currents, for which

\[
\begin{align*}
\mathbf{E}_{\alpha}(r, t) &= 1/2(E_{\omega \alpha}(r) e^{-i\omega t} + E^{*}_{\omega \alpha}(r) e^{i\omega t}) \\
\mathbf{j}_{\alpha}(r, t) &= 1/2(j_{\omega \alpha}(r) e^{-i\omega t} + j^{*}_{\omega \alpha}(r) e^{i\omega t})
\end{align*}
\]

The expression

\[
\Pi_{\omega\alpha\beta}(\delta_{\alpha\beta} - k_{\alpha}k_{\beta}/k^2)
\]

is the operation of projecting on the directions perpendicular to \( k \); it singles out only the transverse fields. This corresponds to the fact that the magnetization current is determined only by the transverse (vortex) component of the electric field. Formula (1.5.11) implies an expression for the contribution to \( \chi^{(1)}_{m}(k, \omega) \) due to magnetization currents. Taking into account (1.5.3) and (1.5.6), we derive from (1.5.11) that

\[
\chi^{(1)}_{m}(k, \omega) = \frac{\varepsilon_{m}^{2}k_{m}^{2}\chi_{m}}{\omega^2} \left( \delta_{\alpha\beta} - \frac{k_{\alpha}k_{\beta}}{k^2} \right) \tag{1.12}
\]

Note that this contribution to the susceptibility \( \chi^{(1)} \) at low frequencies has a singularity \( \omega^{-2} \). The origin of this singularity is purely formal. The corresponding vortex electric field \( \mathbf{E} \sim \partial \mathbf{H}/\partial t \) is very low and is proportional to the frequency so that the formula for susceptibility includes the factor \( \omega^{-1} \). The second factor \( \omega^{-1} \) originates in definition (1.5).
we obtain from (1.6.1) after averaging over time

\[
\overline{Q} = \frac{1}{4V} \int \left( j_{\omega}(r) E_{\omega\alpha}(r) + j^*_{\omega\alpha}(r) E_{\omega\alpha}(r) \right) d^3r
\]

\[
= \frac{\omega}{4V} \int d^3r \int d^3r' \left( \chi^{(1)}_{\alpha\beta}(r, r'; \omega) E_{\omega\alpha}(r) E_{\omega\beta}(r') - \chi^{(1)*}_{\alpha\beta}(r', r; \omega) E_{\omega\beta}(r') \right)
\]

\[
= \frac{\omega}{2V} \int d^3r \int d^3r' E_{\omega\alpha}(r) \frac{\chi^{(1)}_{\alpha\beta}(r, r'; \omega) - \chi^{(1)*}_{\beta\alpha}(r', r; \omega)}{2i} E_{\omega\beta}(r')
\]

(1.6.2)

We have used here the expression (1.4.1) for the current and a relation of the type of (1.5.5) between \(\sigma\) and \(\chi^{(1)}_{\alpha\beta}(r, r'; \omega)\) stand for the Fourier transforms \(\chi^{(1)}_{\alpha\beta}(r, r'; t - t')\) in time. The integration in \(r'\) is carried out both over the volume \(V\) and over \(r\) because it is assumed that if \(|r - r'|\) is greater than the nonlocality radius, then \(\chi^{(1)}_{\alpha\beta}(r, r'; \omega) \approx 0\). At the last step, summation and integration variable in the second term in (1.6.2) have been exchanged: \(\alpha \leftrightarrow \beta\) and \(r \leftrightarrow r'\).

For plane waves, that is,

\[E_{\omega\alpha}(r) = E_{k\omega\alpha} e^{ikr}\]

we obtain for spatially uniform medium, instead of (1.6.2)

\[
\overline{Q} = \frac{\omega}{2} \frac{1}{2i} \left( \chi^{(1)}_{\alpha\beta}(k, \omega) - \chi^{(1)*}_{\beta\alpha}(k, \omega) \right) E_{k\omega\alpha}^* E_{k\omega\beta}
\]

(1.6.3)

If the field \(E\) is taken as a sum of plane waves with different values of \(k\) (a Fourier integral or Fourier series), then integration over a large volume \(V\) yields a sum, or integral, of expressions of the type of (1.6.3) over \(k\).

Expressions (1.6.2) and (1.6.3) show that losses are determined by the anti-Hermitian component of the linear susceptibility tensor

\[
\frac{1}{2i} \left( \chi^{(1)}_{\alpha\beta}(r, r'; \omega) - \chi^{(1)*}_{\beta\alpha}(r', r; \omega) \right)
\]

or, for a spatially uniform medium, by

\[
\frac{1}{2i} \left( \chi^{(1)}_{\alpha\beta}(k, \omega) - \chi^{(1)*}_{\beta\alpha}(k, \omega) \right)
\]

The susceptibilities which we began discussing in section 1.4, are the true susceptibilities that relate polarization and the statistically average field in the medium. These are the relations which complement the system of Maxwell's equations (1.3.1) or (1.3.3) and are introduced in the phenomenological theory.

Another approach is to begin with external fields (in the absence of the medium) or sources \(j^{(e)}\) and \(\rho^{(e)}\) of external fields, as in equations (1.3.2) and (1.3.3). These sources would produce, in the absence of the medium, external fields \(E^{(e)}\), and we can pose the problem of finding the response of the system, that is, the current and charge densities \(j\) and \(\rho\), to the external field. The expansion of the current in powers of external field yields expressions of the same type as (1.4.1), (1.4.3) and (1.4.4), but with \(E\) replaced by \(E^{(e)}\), and \(\sigma\) and \(\chi\) by the responses to the external field, \(\sigma^{(e)}\) and \(\chi^{(e)}\).

The point is that it is \(\chi^{(e)}\) (or \(\sigma^{(e)}\)) and not \(\chi\) that the microscopic theory calculates using quantum mechanics. We will show this to be true in the next chapter when deriving the general expressions for susceptibilities.

In applications, however, one needs the response to the true field, that is, the susceptibility \(\chi\). The problem is to establish a relation between \(\chi\) and \(\chi^{(e)}\), in order to find the response in the medium to the true field from the response to the external field calculated in terms of quantum mechanics.

We begin with Maxwell's equations with external currents, (1.3.2) or (1.3.3). As usual, we can exclude the magnetic field and write equations for the electric field:

\[
\text{curl} \text{curl} E + \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} + 4\pi \frac{\partial^2 \rho}{\partial t^2} = -\frac{4\pi}{c^2} \frac{\partial j^{(e)}}{\partial t}
\]

(1.6.4)
After Fourier transforms of all fields of the type (1.5.1) or (1.5.2) are found, we arrive at Fourier transforms

\[ \mathcal{D}^{-1}_{0\alpha\beta}(k, \omega) E_{k\omega\beta} - \frac{4\pi \omega^2}{c^2} P_{k\omega\alpha} = \frac{4\pi i \omega}{c^2} j^{(e)}_{k\omega\alpha} \]  

(1.6.5)

We have introduced here a tensor

\[ \mathcal{D}^{-1}_{0\alpha\beta}(k, \omega) = \left( k^2 - \frac{\omega^2}{c^2} \right) \delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{k^2} \]  

(1.6.6)

Equation (1.6.5) is obtained from (1.6.4) if we recall that the field

\[ \mathcal{E} = \mathcal{E}^{eikr} + \text{C.C.} \]

(C.C. stands for complex-conjugate expressions) implies that

\[ \text{curl} \mathcal{E} = i[\mathbf{k} \mathcal{E}] \text{e}^{ikr} + \text{C.C.} \]

\[ \text{curl} \text{curl} \mathcal{E} = -[\mathbf{k}[\mathbf{k} \mathcal{E}]] \text{e}^{ikr} + \text{C.C.} = (-k(k \mathcal{E}) + k^2 \mathcal{E}) \text{e}^{ikr} + \text{C.C.} \]

We will need the tensor inverse to \( \mathcal{D}^{-1}_0 \). It is easy to find if we single out in \( \mathcal{D}^{-1}_0 \) its longitudinal and transverse parts. First we recast \( \mathcal{D}^{-1}_0 \) to the form

\[ \mathcal{D}^{-1}_{0\alpha\beta}(k, \omega) = \left( k^2 - \frac{\omega^2}{c^2} \right) \Pi_{\parallel\alpha\beta}(k) - \frac{\omega^2}{k^2} \Pi_{\perp\alpha\beta}(k) \]  

(1.6.7)

where

\[ \Pi_{\parallel\alpha\beta}(k) = \frac{k_\alpha k_\beta}{k^2} \quad \text{and} \quad \Pi_{\perp\alpha\beta}(k) = \delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{k^2} \]

are the operators of projection to the directions which are parallel and perpendicular, respectively, to \( k \).

The projection operators \( \Pi_{\parallel} \) and \( \Pi_{\perp} \) have the following properties:

\[ \Pi_{\perp} \Pi_{\perp} = \Pi_{\perp} \quad \Pi_{\parallel} \Pi_{\parallel} = \Pi_{\parallel} \quad \Pi_{\parallel} \Pi_{\perp} = \Pi_{\perp} \Pi_{\parallel} = 0 \quad \Pi_{\perp} + \Pi_{\parallel} = 1 \]

If an arbitrary operator \( \Gamma \) can be written as

\[ \Gamma = \gamma_{\parallel} \Pi_{\parallel} + \gamma_{\perp} \Pi_{\perp} \]

where \( \gamma_{\parallel} \) and \( \gamma_{\perp} \) are numerical coefficients, then it is not difficult to verify that

\[ \Gamma^{-1} = \frac{1}{\gamma_{\parallel}} \Pi_{\parallel} + \frac{1}{\gamma_{\perp}} \Pi_{\perp} \]

Indeed,

\[ \Gamma^{-1} = (\gamma_{\parallel} \Pi_{\parallel} + \gamma_{\perp} \Pi_{\perp}) \left( \frac{1}{\gamma_{\parallel}} \Pi_{\parallel} + \frac{1}{\gamma_{\perp}} \Pi_{\perp} \right) = \Pi_{\parallel} + \Pi_{\perp} = 1 \]

Equation (1.6.7) implies, therefore, that

\[ \mathcal{D}_0 \equiv (\mathcal{D}_0^{-1})^{-1} = \frac{1}{k^2 - \omega^2/c^2} \Pi_{\perp} - \frac{\omega^2}{k^2} \Pi_{\parallel} \]  

(1.6.8)

By multiplying equation (1.6.5) by \( \mathcal{D}_0 \), we obtain

\[ (1 - \frac{4\pi \omega^2}{c^2} \mathcal{D}_0 \chi^{(1)}) \mathcal{E} = \frac{4\pi i \omega}{c^3} \mathcal{D}_0 \mathcal{E} \]  

(1.6.9)

For the sake of brevity, we have dropped the tensor suffixes \( \alpha, \beta \) and made use of the matrix notation.

Assuming \( \chi^{(1)} = 0 \) in (1.6.9), we obtain an expression for the external field (the field produced by the current \( j^{(e)} \) in the absence of matter):

\[ \mathcal{E}^{(e)} = \frac{4\pi i \omega}{c^3} \mathcal{D}_0 j^{(e)} \]  

(1.6.10)

Equation (1.6.9) gives

\[ (1 - \frac{4\pi \omega^2}{c^2} \mathcal{D}_0 \chi^{(1)}) \mathcal{E} = \mathcal{E}^{(e)} \]  

(1.6.11)

that is,

\[ \mathcal{E} = \left(1 - \frac{4\pi \omega^2}{c^2} \mathcal{D}_0 \chi^{(1)}\right)^{-1} \mathcal{E}^{(e)} \]  

(1.6.12)

Formulas (1.6.11) and (1.6.12) express the external field in terms of the true field, and vice versa.

The definition

\[ P^{(1)} = \chi^{(1)} \mathcal{E} = \chi^{(1)\mathcal{E}} \]  

and equation (1.6.12) imply that

\[ \chi^{(1)\mathcal{E}} = \chi^{(1)} \left(1 - \frac{4\pi \omega^2}{c^2} \mathcal{D}_0 \chi^{(1)}\right)^{-1} \]  

(1.6.13)

This formula expresses \( \chi^{(1)\mathcal{E}} \) in terms of \( \chi^{(1)} \), although the reverse relation is more interesting. Equation (1.6.13) is readily solved for \( \chi^{(1)} \):

\[ \chi^{(1)} = \chi^{(1)\mathcal{E}} \left(1 - \frac{4\pi \omega^2}{c^2} \mathcal{D}_0 \chi^{(1)}\right) = \chi^{(1)\mathcal{E}} - \frac{4\pi \omega^2}{c^2} \chi^{(1)\mathcal{E}} \mathcal{D}_0 \chi^{(1)} \]

Transferring the last term of the righthand side to the left, we obtain

\[ \left(1 + \frac{4\pi \omega^2}{c^2} \chi^{(1)\mathcal{E}} \mathcal{D}_0\right) \chi^{(1)} = \chi^{(1)\mathcal{E}} \]
that is,
\[ \chi^{(4)} = \left(1 + \frac{4 \pi \omega^2}{c^2} \chi^{(1e)} D_0\right)^{-1} \chi^{(1e)} \]  
(1.6.14)

The identity
\[ \left(1 + \frac{4 \pi \omega^2}{c^2} \chi^{(1e)} D_0\right) \chi^{(1e)} = \chi^{(1e)} \left(1 + \frac{4 \pi \omega^2}{c^2} D_0 \chi^{(1e)}\right) \]

which implies
\[ \left(1 + \frac{4 \pi \omega^2}{c^2} \chi^{(1e)} D_0\right)^{-1} \chi^{(1e)} = \chi^{(1e)} \left(1 + \frac{4 \pi \omega^2}{c^2} D_0 \chi^{(1e)}\right)^{-1} \]

enables us to rewrite (1.6.14) in an equivalent form:
\[ \chi^{(1)} = \chi^{(1e)} \left(1 + \frac{4 \pi \omega^2}{c^2} D_0 \chi^{(1e)}\right)^{-1} \]  
(1.6.15)

Formulas (1.6.14) and (1.6.15) make it possible to find expressions for \( \chi^{(1)} \) once the susceptibilities for the external field \( \chi^{(1e)} \) have been found from the microscopic theory.

Using expressions (1.6.11)–(1.6.15), we can express the losses in terms of the external fields and the susceptibilities \( \chi^{(1e)} \). Equation (1.6.11) implies that
\[
\frac{1}{2i} \left( E^{(s)+} - \frac{4 \pi \omega^2}{c^2} \chi^{(1e)} \right) E^{(s)} = \frac{1}{2i} \left( E^{(s)+} - \frac{4 \pi \omega^2}{c^2} \chi^{(1e)} \right) E^{(s)} + \frac{1}{2i} \left( E^{(s)+} - \frac{4 \pi \omega^2}{c^2} \chi^{(1e)} \right) E^{(s)}
\]

(1.6.16)

(imaginary part of susceptibility, or in terms of the external field and the imaginary part of the response to the external field. The physical meaning expressing the losses via the external field is that the fields \( E \) and \( E^{(s)} \) differ by fields that are generated by the induced currents. The induced field then describes the internal interaction between particles of a medium and does not lead to dissipation. What really dissipates is the energy of the external field.

1.7. Nonlinear Susceptibilities

The retardation effect and nonlocality (and, correspondingly, dispersion and spatial dispersion) characterize nonlinear susceptibilities as well as linear ones.

The general relation of the current density \( j \) and the field \( E \) in the second order in field is given by (1.4.2). The integration in time is carried out from \( -\infty \) to \( t \), and that in coordinates, over the entire space.

In a stationary and spatially uniform medium we have
\[
\rho^{(2)}_{\alpha\gamma} = \rho^{(2)}_{\alpha\gamma}(r - r', r - r'\prime; t - t', t - t'\prime)
\]

It will again be more convenient to transfer to the Fourier representation (1.5.1) and (1.5.2). Let us introduce the notation
\[
R' = r - r' \quad R'' = r - r'' \quad t' = t - t' \quad t'' = t - t''
\]

The quantities \( r' \) and \( r'' \) are the response delay times at the moment of time \( t \) relative to the fields acting at the moments \( t' \) and \( t'' \). By virtue of the causality principle, \( t' \geq 0 \) and \( t'' \geq 0 \).

If (1.4.2) is multiplied by \( \text{exp}(i\omega \cdot t - ikr) \) and integrated over \( t \) and \( r \), we obtain
\[
j^{(2)}_{\omega\alpha} = \int dt \int d^3r \int d^3r' \int d^3r'' \int d^3R' \int d^3R'' \rho^{(2)}_{\alpha\gamma}(R', R'', t', t'')
\]

(1.6.17)

Integration in \( t \) and \( r \) can now be carried out; using then the integral representation of the \( \delta \) function
\[
(2\pi)^{3}\delta(w - \omega' - \omega'')\delta^3(k - k' - k'') = \int e^{-i(k-k'k'k'')} t dt d^3r
\]

The dissipation of the energy of the field in a medium thus be given by two equivalent formulas: either in terms of the mean field and the
we find
\[ j_{\omega,\alpha}^{(2)} = \frac{\int d^3 k' d^3 k'' \, d\omega' \, d\omega''}{(2\pi)^4} \delta^3(k - k' - k'') \delta(\omega - \omega' - \omega'') \times \sigma_{\alpha\beta\gamma}^{(2)}(k', k''; \omega', \omega'') E_{k' \omega' \beta} E_{k'' \omega'' \gamma} \]
(1.7.1)
where
\[ \sigma_{\alpha\beta\gamma}^{(2)}(k', k''; \omega', \omega'') = \int_0^\infty d\tau'' \int d^3 R' d^3 R'' \sigma_{\alpha\beta\gamma}^{(2)}(R', R''; \tau', \tau'') \times \exp(-ik'R - ik''R'' + i\omega'\tau' + i\omega''\tau'') \]
is the Fourier transform of the function \( \sigma_{\alpha\beta\gamma}^{(2)} \).

The relation of the current \( j_{\omega,\alpha}^{(3)} \) to the fields is not algebraic here, but integral. However, if only two plane harmonic modes of the field, \( k'\omega' \) and \( k''\omega'' \), are nonzero, it produces a current
\[ j_{k' + k'', \omega + \omega', \alpha}^{(2)} = \sigma_{\alpha\beta\gamma}^{(2)} E_{k' \omega' \beta} E_{k'' \omega'' \gamma} \]

As in the linear case, not the conductivity \( \sigma^{(2)} \) but susceptibility \( \chi^{(2)} \) is often used. Their relationship is similar to (1.5.5):
\[ \chi_{\alpha\beta\gamma}^{(2)}(k = k' + k''; \omega = \omega' + \omega'') = \frac{i}{\omega} \sigma_{\alpha\beta\gamma}^{(2)}(k = k' + k''; \omega = \omega' + \omega'') \]
(1.7.2)

Owing to \( \delta \) functions, (1.7.1) can be integrated, for example, in \( k'' \) and \( \omega'' \) but it is more convenient to retain the form of (1.7.1) in which \( \delta \) functions indicate the condition of summation of frequencies and the synchronism condition:
\[ \omega = \omega' + \omega'' \quad k = k' + k'' \]
(1.7.3)
The situation with cubic susceptibility and higher-order susceptibilities is quite similar.

Let us see what form the loss expressions will take if the quadratic contribution to current is taken into account. We assume that there are three waves which satisfy conditions (1.7.3). For simplicity, we neglect spatial dispersion. By analogy to (1.6.3), we obtain the following expression for the losses at a frequency \( \omega \), caused by the quadratic current \( j_{\omega,\alpha}^{(3)} \):
\[ \overline{Q}_{\omega} = J_{\alpha}(r, t) E_{\alpha}(r, t) \]
\[ = \frac{\varepsilon}{2i} \left( \chi_{\alpha\beta\gamma}^{(2)} E_{\omega\alpha}^{\star} E_{\omega\beta} E_{\omega''\gamma}^{\star} - \chi_{\alpha\beta\gamma}^{(2)} E_{\omega\alpha} E_{\omega'\beta}^{\star} E_{\omega''\gamma} \right) \]
\[ = \frac{\varepsilon}{2} \text{Im}(\chi_{\alpha\beta\gamma}^{(2)}(\omega = \omega' + \omega'') E_{\omega\alpha}^{\star} E_{\omega\beta} E_{\omega''\gamma}^{\star}) \]
(1.7.4)

The averaging is carried out over time, so that rapidly oscillating terms in \( Q \) are eliminated.

In the linear case, the losses were expressed in terms of the imaginary part of susceptibility; here, they are found in terms of the imaginary part of the product of susceptibility \( \chi^{(2)} \) by the amplitudes of the fields. These amplitudes are complex so that the losses depend on the relative phases of the fields at the frequencies \( \omega, \omega' \) and \( \omega'' \).

The physical meaning of this result is that only one wave at the frequency \( \omega \) is produced in the linear case, and the polarization arises at the same frequency. Losses depend on the relative phases of current and field (or polarization and field), and this relation between phases is determined by the phase of susceptibility. In the nonlinear case, however, there are three fields, and the polarization at the frequency \( \omega \) is produced by the fields at the frequencies \( \omega' \) and \( \omega'' \), so that the losses depend on the relative phases of all three fields.

In some cases, losses may be negative, that is, there may be amplification at the frequency \( \omega \). Negative losses may simply indicate a transfer of the field energy from some modes to others. Therefore, formula (1.7.4) covers both the true losses and energy redistribution.

If the medium is transparent at all three frequencies (there is no real dissipation of energy), then energy changes in each of the modes can only be caused by energy transfer to other modes. Therefore,
\[ \overline{Q}_{\omega} + \overline{Q}_{\omega'} + \overline{Q}_{\omega''} = 0 \]
This leads to certain constraints on the tensor \( \chi^{(2)} \). To find them, we use (1.7.4) and similar expressions for \( \overline{Q}_{\omega'} \) and \( \overline{Q}_{\omega''} \) and arrive at
\[ \frac{\omega}{2} \text{Im}(\chi_{\alpha\beta\gamma}^{(2)}(\omega = \omega' + \omega'') E_{\omega\alpha}^{\star} E_{\omega'\beta} E_{\omega''\gamma}) + \frac{\omega'}{2} \text{Im}(\chi_{\alpha\beta\gamma}^{(2)}(\omega' = \omega - \omega'') E_{\omega\alpha}^{\star} E_{\omega''\beta} E_{\omega'\gamma}) = 0 \]
(1.7.5)
The summation indices \( \alpha, \beta \) and \( \gamma \) in this expression can be rearranged so that each term will contain an identical product of fields, \( E_{\omega\alpha}^{\star} E_{\omega'\beta} E_{\omega''\gamma} \). Equality (1.7.5) must hold for any value of amplitudes of the fields, which requires that
\[ \omega \chi_{\alpha\beta\gamma}^{(2)}(\omega = \omega' + \omega'') - \omega' \chi_{\beta\alpha\gamma}^{(2)}(\omega = \omega - \omega'') - \omega'' \chi_{\gamma\alpha\beta}^{(2)}(\omega'' = \omega - \omega') = 0 \]
(1.7.6)

It will be shown later that if there is no true dissipation, that is, if we are far from absorption bands, the susceptibility is a real quantity. We can
assume, therefore, that instead of imposing the limitation (1.7.6), we can choose stronger symmetry constraints:

$$\chi_{\alpha\beta\gamma}^{(2)}(\omega = \omega' + \omega'') = \chi_{\beta\gamma\delta}^{(2)}(\omega = \omega' - \omega'') = \chi_{\gamma\delta\alpha}^{(2)}(\omega'' = \omega - \omega')$$  \hspace{1cm} (1.7.7)

These symmetry relations can be formulated into a rule: when rearranging frequencies in $\chi^{(2)}$, also rearrange the corresponding polarization indices. This can be shown by the diagram in Figure 2.2 (see Chapter 2).

The significance of this result lies in the fact that having measured $\chi^{(2)}$ for one of the processes, say, addition of frequencies $\omega = \omega' + \omega''$, we can at the same time determine $\chi^{(2)}$ for the processes of frequency subtraction, that is, $\omega'' = \omega - \omega'$. This implies

$\chi_{\alpha\beta\gamma}^{(2)}(\omega = \omega' - \omega'') = \chi_{\beta\gamma\delta}^{(2)}(\omega = \omega' + \omega'') = \chi_{\gamma\delta\alpha}^{(2)}(\omega' = \omega - \omega')$.

Relation (1.7.7) will be given rigorous proof in the next chapter, using an explicit expression for quadratic susceptibilities.

Similar arguments hold also for the cubic susceptibility and higher-order susceptibilities. The difference lies only in that in the case of the cubic susceptibility and odd-order susceptibilities in general, degenerate cases are possible, when losses are expressed in terms of the imaginary component of a susceptibility.

Among all other processes described by $\chi_{\alpha\beta\gamma\delta}^{(3)}(\omega = \omega' + \omega'' + \omega''')$, there are special cases when, for instance, $\omega'' = -\omega'$. Since the field is real, a field component at a frequency $-\omega'$ is always present in addition to the component at a frequency $\omega'$. This implies

$$E_{\omega''\gamma} = E_{\omega'\gamma}^*$$

By analogy to (1.7.4), losses are found from the expression

$$Q_{\omega} = \frac{\omega}{2} \text{Im}(\chi_{\alpha\beta\gamma\delta}^{(3)}(\omega = \omega' - \omega' + \omega)E_{\omega\alpha}E_{\omega\beta}E_{\omega\gamma}^*E_{\omega\delta})$$

and are thus essentially independent of the phase of the field at the frequencies $\omega$ and $\omega'$.

In this case, losses are determined by the imaginary part of the cubic susceptibility and correspond to the two-photon absorption or two-photon scattering.

1.8. Susceptibilities of a Crystal

The results presented in Sections 1.5–1.7 mostly treated spatially uniform media. We have already mentioned that crystals must be treated as a special case. Classical optics of crystals uses averaging over a physically infinitely small volume of the medium, which contains a large number of particles; for crystals, this means a volume whose dimensions are much greater than $a_0$ or the interatomic distance. After this averaging, a crystal can be treated as a spatially uniform medium, which substantially simplifies the solution of problems in crystal optics.

We have indicated, however, that the procedure of averaging over a physically infinitely small volume (small in comparison with the wavelength) cannot be used for the short-wavelength electromagnetic field (the x-ray range) and that even in the optical range this averaging eliminates a number of principally important effects.

If, however, we forget the averaging over volume and if we restrict the averaging to statistical averaging over the Gibbs distribution, we come across certain mathematical complications caused by inherent nonuniformity of crystals. By definition, a crystal is a periodic nonuniform medium. The periodicity is imposed by the Bravais lattice of the crystal,

$$R_n = n_1a_1 + n_2a_2 + n_3a_3$$

where $R_n$ is an arbitrary vector of the Bravais lattice, $a_1, a_2, a_3$ are the basis vectors of the lattice and $n_1, n_2, n_3$ are integers composing an integral-valued vector $n$.

An arbitrary scalar function $\rho(r)$, which is periodical on the Bravais lattice, has the property

$$\rho(r + R_n) = \rho(r)$$  \hspace{1cm} (1.8.1)

One example of such a function can be the mean electron density in the crystal. If $\rho(r)$ is expanded in a Fourier series,

$$\rho(r) = \sum_k \rho_k e^{ikr}$$

then condition (1.8.1) implies that the equality

$$e^{ikR_n} = 1$$

holds for an arbitrary vector of the Bravais lattice, that is,

$$kR_n = 2\pi M$$  \hspace{1cm} (1.8.2)

where $M = 0, \pm 1, \pm 2\ldots$ is an integer. If $k_1$ and $k_2$ satisfy condition (1.8.2), their linear combination with integral coefficients also satisfies (1.8.2). Hence, all vectors $k$ satisfying (1.8.2) make up a lattice known as the reciprocal lattice of the crystal.

The basis vectors of the reciprocal lattice can be written in the form

$$g_1 = \frac{2\pi[a_2a_3]}{(a_1a_2a_3)} \quad g_2 = \frac{2\pi[a_3a_1]}{(a_1a_2a_3)} \quad g_3 = \frac{2\pi[a_1a_2]}{(a_1a_2a_3)}$$