

Chapter 2

Interaction of Ultrashort Electromagnetic Pulses with Matter: Description in the Framework of Perturbation Theory

Considerable advances have been made in the generation of ultrashort electromagnetic field pulses of controlled shape over a wide spectral range [1]. In the infrared, visible, and far-ultraviolet spectral regions, pulses have been produced with widths equal to the period of the electromagnetic field oscillation at the carrier frequency (single-cycle pulses). Single-cycle pulses have widths of a few femtoseconds in the near-infrared and visible regions and of the order of a hundred attoseconds and less in the far-UV range. Such pulses provide the basis for studying electron dynamics with resolutions approaching one atomic time unit (24 as). In the terahertz range, half-cycle pulses are generated with widths of the order of a picosecond, which is promising in particular for quantum calculations using Rydberg states [2].

With the development of techniques for generating ultrashort pulses of electromagnetic radiation, it has become urgent to consider the peculiar features of interactions between radiation and specific substances, and to develop adequate ways of describing electromagnetic processes in ultrashort fields. The use of ultrashort laser pulses opens up new possibilities in superfast monitoring of light-induced phenomena, attosecond metrology, spectroscopy, microscopy, and plasmonics.

The interaction between single-cycle and subcycle pulses and matter has characteristic features that differ from the case of multicycle pulses. For the latter, it is well known that the photoprocess probability does not depend on the phase of the electromagnetic field, but is determined by the strength and carrier frequency. An important peculiarity of ultrashort interactions is the dependence of the photoprocess probability on the phase characteristics of the radiation, and in particular the carrier phase with respect to the envelope and the frequency chirp. (The frequency chirp in the linear case is a coefficient determining the time dependence of the frequency.) This phase dependence can be used both to obtain information about wave functions of atomic electrons and also to use the phase method to control light-induced processes.

In the previous chapter we considered the elementary case where the interaction of a substance with ultrashort pulses can be described by a classical oscillator. Hereafter (Chaps. 2 and 3), these interactions will be investigated in the framework

of a more realistic approach, the semiclassical theory, where the substance is described at the quantum level and the radiation is described classically.

In this chapter it is assumed that the electric field in the ultrashort pulse is not too high (or the photoprocess cross-section is small enough) to justify applying the quantum–mechanical perturbation theory.

2.1 Derivation of the Basic Formula

When perturbation theory is applicable and one considers the action of relatively long radiation pulses on a substance, the response to the electromagnetic action is usually described with reference to the photoprocess probability per unit time w .

This can be determined from the cross-section σ of the photoprocess by the formula

$$w = \int \sigma(\omega') \frac{I(\omega')}{\hbar \omega'} d\omega', \quad (2.1)$$

where $I(\omega')$ is the spectral intensity of radiation which, for a monochromatic field of frequency ω , is $I(\omega') = I_0 \delta(\omega - \omega')$, with I_0 the integrated intensity.

It should be noted that the cross-section $\sigma(\omega')$ corresponds to the action of a *monochromatic* field at a specified frequency ω' , while the integration over frequencies on the right-hand side of (2.1) takes into account the non-monochromaticity of the radiation. If the pulse lasts much longer than the period of oscillation $T = 2\pi/\omega$ at the carrier frequency, the spectral intensity of radiation is determined in terms of the electric field strength autocorrelator by the formula

$$I(\omega') = \frac{c}{(2\pi)^2} \int_{-\infty}^{\infty} \langle E_i(t) E_i(t + \tau) \rangle_t \exp(i\omega' \tau) d\tau, \quad (2.2)$$

where the symbol $\langle \dots \rangle_t$ denotes the time average (see the derivation of (2.2) in Appendix 1 and note that $I(\omega') = c \rho(\omega')$).

In going to ultrashort pulses consisting of a small number of optical cycles or even of part of a cycle, the concept of photoprocess probability per unit time becomes inadequate, and a description of the electromagnetic interaction through the total probability for the whole time of action of the pulse is more physically meaningful. So for ultrashort pulses, the formulas (2.1)–(2.2) are no longer valid.

To describe processes in ultrashort fields within the framework of the perturbation theory, (2.1) is replaced by a calculation of the *total* photoprocess probability W for the whole time of action of the ultrashort pulse, retaining the description of the properties of the excited system in terms of the cross-section $\sigma(\omega')$. This is the problem to which this section is dedicated.

So let us consider photoexcitation of a quantum system from the ground state $|0\rangle$ to some excited state $|n\rangle$ under the action of a dipole perturbation

$$\hat{V}(t) = -\hat{\mathbf{d}}_i E_i(t), \quad (2.3)$$

where $\hat{\mathbf{d}}$ is the electric dipole moment operator of the system and $\mathbf{E}(t)$ is the electric field strength, taken to be a classical quantity that is independent of the spatial coordinate (dipole approximation). To first order in perturbation theory, the amplitude of this process for the whole time of action of the field is

$$c_{n0} = -\frac{i}{\hbar} \int_{-\infty}^{\infty} \langle n | \hat{d}_i(t) | 0 \rangle E_i(t) dt, \quad (2.4)$$

where $\hat{d}_i(t) = \exp(i\hat{H}_0 t/\hbar) \hat{d}_i \exp(-i\hat{H}_0 t/\hbar)$ is the dipole moment operator in the interaction representation and \hat{H}_0 is the zero-order Hamiltonian of the system.

The probability of photoexcitation with the transition $|0\rangle \rightarrow |n\rangle$ for the whole time of action of the perturbation is

$$W_{n0} = \frac{1}{\hbar^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \langle 0 | \hat{d}_i(t') | n \rangle \langle n | \hat{d}_k(t) | 0 \rangle E_i(t') E_k(t) dt dt'. \quad (2.5)$$

The total photoexcitation probability taking into account transitions to all states of the system is then

$$W_{tot} = \sum_n W_{n0} = \frac{1}{\hbar^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \langle 0 | \hat{d}_i(t') \hat{d}_k(t) | 0 \rangle E_i(t') E_k(t) dt dt'. \quad (2.6)$$

In the derivation of (2.6) from (2.5), we used the completeness of the set of functions $|n\rangle$, and the fact that $\langle 0 | \hat{d}_i(t) | 0 \rangle = 0$ for a spherically symmetrical system, so to satisfy the completeness condition in the sum (2.6), a summand can be included that corresponds to the invariable state of the system W_{00} .

We now use the fact that the correlator of dipole moments of a system that is stationary in the unperturbed state depends only on the time difference $\tau = t' - t$: $K_{ik}(\tau) = \langle 0 | \hat{d}_i(t') \hat{d}_k(t) | 0 \rangle$, something that is easy to check directly. Then the formula (2.6) can be rewritten as

$$W_{tot} = \frac{1}{\hbar^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} K(\tau) E_i(t + \tau) E_i(t) dt d\tau. \quad (2.7)$$

Here we note that, for a centrosymmetric system, $K_{ik}(\tau) = \delta_{ik} K(\tau)$, where $K(\tau) = K_{ii}(\tau)/3$. Replacing the terms in the integrand of (2.7) by Fourier transforms $f(t) = \int_{-\infty}^{\infty} f(\omega') \exp(i\omega' t) \frac{d\omega'}{2\pi}$, we obtain

$$W_{tot} = \frac{2\pi}{\hbar^2 c} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} K(\omega') I(\omega', t) d\omega' dt, \quad (2.8)$$

where we have introduced the function

$$I(\omega', t) = \frac{c}{(2\pi)^2} \int_{-\infty}^{\infty} E_i(t) E_i(t + \tau) \exp(i\omega' \tau) d\tau. \quad (2.9)$$

This function can be called the spectral density of instantaneous radiation intensity. The Fourier transform of the dipole moment correlator is by definition

$$K(\omega) = \frac{1}{3} \int_{-\infty}^{\infty} \langle 0 | \hat{d}_i(t) \hat{d}_i(t + \tau) | 0 \rangle \exp(i\omega \tau) d\tau. \quad (2.10)$$

Starting from (2.9), it is not difficult to prove that

$$\int_{-\infty}^{\infty} I(\omega', t) dt = \frac{c}{(2\pi)^2} |\mathbf{E}(\omega')|^2. \quad (2.11)$$

Substituting the expression (2.11) into the right-hand side of (2.8), we obtain

$$W_{tot} = \frac{1}{2\pi\hbar^2} \int_{-\infty}^{\infty} K(\omega') |\mathbf{E}(\omega')|^2 d\omega'. \quad (2.12)$$

Let us apply this to a monochromatic field $\mathbf{E}(t) = \mathbf{E}_0 \cos(\omega t)$, where the average intensity over the period $T = 2\pi/\omega$ is

$$\langle I(\omega', t) \rangle_T = I_0 \delta(\omega - \omega') \langle 1 + \exp(-2i\omega t) \rangle_T = I_0 \delta(\omega - \omega'), \quad (2.13)$$

with $I_0 = c\mathbf{E}_0^2/8\pi$. Integrating (2.13) over the finite time interval Δt as in (2.8), we find

$$W_{tot} = \frac{2\pi}{\hbar^2 c} K(\omega) I_0 \Delta t, \quad (2.14)$$

whence the photoexcitation probability *per unit time* is

$$w_{tot} = \frac{2\pi}{\hbar^2 c} K(\omega) I_0. \quad (2.15)$$

On the other hand, by definition of the photoabsorption cross-section in a monochromatic field of frequency ω , we have

$$w_{tot} = \sigma(\omega) \frac{I_0}{\hbar \omega}. \quad (2.16)$$

The value $I_0/\hbar\omega$ represents the monochromatic radiation photon flux density. Comparing (2.15) and (2.16), we find the expression for the Fourier transform of the *dipole moment correlator* in terms of the *photoabsorption cross-section* in the dipole approximation:

$$K(\omega) = \frac{\hbar c}{2\pi\omega} \sigma(\omega). \quad (2.17)$$

This implies the important formula for the photoabsorption cross-section:

$$\sigma(\omega) = \frac{2\pi\omega}{\hbar c} K(\omega). \quad (2.18)$$

Substituting (2.17) in (2.12), we obtain the expression for the total probability of the process under consideration for the whole time of action of the radiation pulse. Since the case in point is photoabsorption, the integration in (2.12) is necessarily restricted to positive frequencies, so finally we find

$$W_{tot} = \frac{c}{(2\pi)^2} \int_0^\infty \sigma(\omega') \frac{|\mathbf{E}(\omega')|^2}{\hbar\omega'} d\omega'. \quad (2.19)$$

Obtained within the framework of the perturbation theory, the expression (2.19) is the *basic formula* describing the total photoprocess probability under the action of ultrashort pulses. Of course, it is true whenever $W_{tot} < 1$, according to the mathematical definition of probability.

For the photoabsorption cross-section the equation can be written

$$\sigma(\omega') = \frac{2\pi^2 e^2}{mc} \left\{ \sum_{n>0} f_{n0} G_{n0}(\omega') + g_{c0}(\omega') \right\} = \sum_n \sigma_{n0}(\omega') + \sigma_{c0}(\omega'), \quad (2.20)$$

where the contributions to the total cross-section of the discrete spectrum (the sum over n) and the continuous spectrum (the second summand in the braces) have been separated. Here f_{n0} are the oscillator strengths, $G_{n0}(\omega')$ is the spectral shape of the line for the transition $|0\rangle \rightarrow |n\rangle$, and $g_{c0}(\omega')$ is the spectral function of dipole excitation to the continuous energy spectrum. The right-hand side of (2.20) involves $\sigma_{n0}(\omega')$, the cross-section for photoexcitation of the discrete power level, and $\sigma_{c0}(\omega')$, the cross-section for excitation of a state in the continuous spectrum. In view of (2.20), the following expression for the total probability of photoexcitation of the n th state of the discrete spectrum can be obtained from (2.19):

$$W_{n0} = \frac{c}{(2\pi)^2} \int_0^\infty \sigma_{n0}(\omega') \frac{|\mathbf{E}(\omega')|^2}{\hbar\omega'} d\omega' \quad (2.21)$$

or

$$W_{n0} = \frac{e^2}{2m} f_{n0} \int_0^\infty G_{n0}(\omega') \frac{|\mathbf{E}(\omega')|^2}{\hbar \omega'} d\omega'. \quad (2.22)$$

Equations (2.21) and (2.22) are valid within the range of applicability of the perturbation theory, when $W_{n0} < 1$.

When the spectral pulse width $\Delta\omega \propto 1/\Delta t$ is much greater than the spectral width of the bound-bound transition $\Delta\omega \gg \Delta\omega_{n0}$, the line shape function can be replaced by the delta function $G_{n0}(\omega') \rightarrow \delta(\omega_{n0} - \omega')$. With this replacement, and in view of the expression for the oscillator strength $f_{n0} = 2m\omega_{n0} |\mathbf{d}_{n0}|^2 / 3\hbar e^2$ (2.21) yields a simple expression for the phototransition probability:

$$W_{n0} = |\Omega(\omega_{n0})|^2, \quad (2.23)$$

where $\Omega(\omega_{n0}) = \mathbf{d}_{n0} \mathbf{E}(\omega_{n0})/\hbar$ is the Fourier transform of the instantaneous Rabi frequency calculated at the eigenfrequency of the excited transition. Naturally, (2.23) can be obtained directly from the electric field strength to first order of the perturbation theory using (2.4) and (2.5).

For the probability of excitation of an arbitrary state in the continuous spectrum over the whole time of action of a field pulse, (2.19) and (2.20) imply

$$W_{c0} = \frac{c}{(2\pi)^2} \int_{I_P/\hbar}^\infty \sigma_{c0}(\omega') \frac{|\mathbf{E}(\omega')|^2}{\hbar \omega'} d\omega', \quad (2.24)$$

where I_P is the ionization potential of the system under consideration. For the differential probability of photoexcitation to a state of the continuous spectrum with energy ε , (2.24) yields

$$\frac{dW}{d\omega_{\varepsilon 0}} = \frac{c}{(2\pi)^2} \sigma_{c0}(\omega_{\varepsilon 0}) \frac{|\mathbf{E}(\omega_{\varepsilon 0})|^2}{\hbar \omega_{\varepsilon 0}}, \quad (2.25)$$

where $\omega_{\varepsilon 0} = (I_P + \varepsilon)/\hbar$ is the frequency of transition to a specified state of the continuous spectrum. The formula (2.25) is obtained from (2.24) by substituting the delta function $\delta(\omega_{\varepsilon 0} - \omega')$ into the integrand on the right-hand side of (2.24). This substitution separates out the transition of the system from the ground state to a state of the continuous spectrum with a specified energy $|0\rangle \rightarrow |\varepsilon\rangle$.

For an *arbitrary* photoinduced process in a quasi-monochromatic field, one can obtain the following expression for the probability per unit time in terms of the process cross-section:

$$w_{tot}(t) = \int_0^{\infty} \sigma(\omega') \frac{I(\omega', t)}{\hbar \omega'} d\omega'. \quad (2.26)$$

Here the cross-section $\sigma(\omega')$ can describe not only photoabsorption, but also other photoprocesses, such as scattering, stimulated bremsstrahlung, and stimulated photorecombination. Integrating (2.26) with respect to time, the probability for the whole time of action of the pulse is

$$W = \int_{-\infty}^{\infty} w(t) dt = \int_0^{\infty} \sigma(\omega') \int_{-\infty}^{\infty} \frac{I(\omega', t)}{\hbar \omega'} dt d\omega'. \quad (2.27)$$

Hence in view of (2.11), we arrive at the formula (2.19).

For a quasi-monochromatic field, analogously to (2.21)–(2.25), the expression for the electromagnetic pulse energy absorbed by the quantum system over the whole time of interaction with the radiation is

$$\Delta E = \int_{-\infty}^{\infty} Q(t) dt = \int_0^{\infty} \sigma(\omega') \int_{-\infty}^{\infty} I(\omega', t) dt d\omega'. \quad (2.28)$$

Hence in view of (2.11), we find

$$\Delta E = \frac{c}{(2\pi)^2} \int_0^{\infty} \sigma(\omega') |\mathbf{E}(\omega')|^2 d\omega'. \quad (2.29)$$

Thus (2.19) and (2.25) can be used to describe not only photoabsorption in an ultrashort pulse, but also other photoprocesses, such as radiation scattering and the stimulated bremsstrahlung effect. For this purpose, a suitable cross-section must be substituted in the right-hand side of (2.19).

Let us consider photoexcitation of the system under the action of a laser pulse with a Gaussian envelope i.e., with electric field strength

$$\mathbf{E}(t) = \mathbf{E}_0 \exp(-t^2/\Delta t^2) \cos(\omega t + \varphi), \quad (2.30)$$

where \mathbf{E}_0 is the amplitude, ω is the carrier frequency, φ is the CE phase, and Δt is the parameter proportional to the pulse duration Δt_p .

The Fourier transform of the field (2.30) has the form

$$E(\omega') = E_0 \frac{\sqrt{\pi}}{2} \Delta t \left\{ \exp \left[-i\phi - \frac{(\omega - \omega')^2 \Delta t^2}{4} \right] + \exp \left[i\phi - \frac{(\omega + \omega')^2 \Delta t^2}{4} \right] \right\}. \quad (2.31)$$

It is natural to define the pulse duration Δt_p as the ratio of the probability W for the whole time of action of the pulse to the probability per unit time w for the same pulse shape in the limit of long duration ($\Delta t_p \gg \omega^{-1}$):

$$\Delta t_p \leftarrow \frac{W}{w}. \quad (2.32)$$

Then for the field (2.30), the relationship between the parameter Δt and the pulse duration Δt_p is

$$\Delta t_p = \sqrt{\frac{\pi}{2}} \Delta t \cong 1.253 \Delta t. \quad (2.33)$$

To derive (2.33), we took the limit $\Delta t \rightarrow \infty$ in the Fourier transform of the pulse (2.31): $|\mathbf{E}(\omega')|^2 \rightarrow (\pi/2)^{3/2} \mathbf{E}_0^2 \Delta t \delta(\omega - \omega')$, whence (2.12) leads to $W_{tot} \rightarrow \sqrt{\pi/2} (K(\omega)/4\hbar^2) \mathbf{E}_0^2 \Delta t$. Comparing the last relation with the expression $w = (K(\omega)/4\hbar^2) \mathbf{E}_0^2$ for the photoexcitation rate in a monochromatic field, (2.32) then implies (2.33).

We now consider the number n_c of cycles in a pulse:

$$n_c = \frac{\Delta t_p}{T} = \frac{\omega \Delta t_p}{2\pi}, \quad (2.34)$$

where $T = 2\pi/\omega$ is the period of oscillation at the carrier frequency. Using (2.33), we obtain the following expression for the parameter Δt appearing in the formulas (2.30)–(2.31) in terms of the number of cycles in a pulse:

$$\Delta t = \frac{2\sqrt{2}\pi n_c}{\omega}. \quad (2.35)$$

Hereafter we will consider ultrashort pulses, for which the number of cycles at the carrier frequency is $n_c \geq 1$ and less.

Expressed in terms of the number of cycles, the squared magnitude of the Fourier transform of the field (2.31), which according to (2.19) defines the photoexcitation probability for the whole time of action of the radiation, has the form

$$|E(\omega')|^2 = 2\pi^2 \left(\frac{n_c E_0}{\omega} \right)^2 G_E(\omega', \omega, n_c) [1 + K_{ph}(\omega', \omega, n_c) \cos(2\varphi)], \quad (2.36)$$

where (compare with (1.36) and (1.37) in Chap. 1)

$$G_E(\omega', \omega, n_c) = \exp \left[-4\pi n_c^2 \left(1 - \frac{\omega'}{\omega} \right)^2 \right] + \exp \left[-4\pi n_c^2 \left(1 + \frac{\omega'}{\omega} \right)^2 \right], \quad (2.37)$$

$$K_{ph}(\omega', \omega, n_c) = \operatorname{sech}\left(8\pi n_c^2 \frac{\omega'}{\omega}\right). \quad (2.38)$$

For the probability of excitation of the discrete power level for the whole time of action of the pulse, (2.22) and (2.36) yield

$$W_{n0} = 2\pi^2 \left(\frac{d_{n0} E_0}{\hbar \omega} n_c\right)^2 G_E(\omega_{n0}, \omega, n_c) [1 + K_{ph}(\omega_{n0}, \omega, n_c) \cos(2\varphi)] \quad (2.39)$$

under the assumption that $W_{n0} < 1$. We see here that the function $G_E(\omega_{n0}, \omega, n_c)$ describes the spectral form of an excitation line of the bound-bound transition, while $K_{ph}(\omega_{n0}, \omega, n_c)$ is the phase modulation factor since it specifies the dependence of the process probability on the CE phase.

Note that, for a pulse of the form (1.26a), instead of the equality (2.37), one should use (1.36a), taking into account the relation (2.35) between the parameters Δt and n_c .

From the resulting expression (2.39) it follows that, under the conditions of validity of the perturbation theory, the dependence of the total probability of photoexcitation of the bound-bound transition under the action of a pulse of the form (2.30) on the CE phase is given by the function $\cos(2\varphi)$. From numerical analysis of the right-hand side of (2.38), it follows that the phase modulation factor has an appreciable value only for subcycle pulses: $n_c < 0.5$. For a fixed value of the parameter n_c , the factor K_{ph} grows with carrier frequency ω , and in this case, according to the expression for the spectral function of excitation (2.37), the process probability decreases.

In the limit of a pulse (2.30) of *zero duration* ($\Delta t \rightarrow 0$), when $E(\omega') \rightarrow \sqrt{\pi} E_0 \Delta t \cos \varphi$, (2.19) takes the form

$$W_{tot} = \frac{c}{4\pi\hbar} (E_0 \Delta t)^2 \cos^2 \varphi \int_0^\infty \frac{\sigma(\omega')}{\omega'} d\omega'. \quad (2.40)$$

Thus in this case the total photoabsorption probability is defined by the frequency integral of the ratio $\sigma(\omega')/\omega'$. In this case the carrier envelope (CE) phase dependence is given by the function $\cos^2 \varphi$, which corresponds to the formula (2.39), since $K_{ph} \rightarrow 1$ as $\Delta t \rightarrow 0$.

Note that, for excitation by a pulse of the form (1.26a), Eq. (2.40) should be rewritten as

$$W_{tot} = \frac{c}{16\pi\hbar} E_0^2 \Delta t^6 \sin^2 \varphi \int_0^\infty \sigma(\omega') \omega'^3 d\omega'. \quad (2.40a)$$

For the total absorbed energy over the whole time of action of the pulse and in the limit $\Delta t \rightarrow 0$, (2.29) implies

$$\Delta E = \frac{\pi}{2} N_e r_e (c \Delta t)^2 E_0^2 \cos^2 \varphi, \quad (2.41)$$

where $r_e = e^2/mc^2$ is the classical electron radius and N_e is the number of electrons in the atom. To derive (2.41), we used the sum rule for the photoabsorption cross-section:

$$\int_0^\infty \sigma(\omega') d\omega' = \frac{2\pi^2 e^2}{mc} N_e. \quad (2.42)$$

Equations (2.40)–(2.41) are valid within the framework of applicability of the perturbation theory, when $W_{tot} < 1$. It follows from (2.19) that the limit of zero pulse duration is realized while satisfying $\Delta t < 1/\Delta\omega_a$, where $\Delta\omega_a$ is the frequency interval giving the main contribution to the process cross-section integrated with respect to the frequency.

Thus the formulas (2.19) and (2.27) derived in this section express the total photoprocess probability for the whole time of action of the radiation in terms of the cross-section of this process and the Fourier transform of the electric field strength in the pulse. The resulting equations describe a photoprocess induced by an ultrashort electromagnetic pulse, when the concepts of probability per unit time and radiation intensity are inapplicable but it is nevertheless possible to use the perturbation theory.

2.2 Excitation of a Substance Under the Action of Ultrashort Pulses

2.2.1 Phase Control of Photoexcitation by an Ultrashort Laser Pulse

We now use (2.19) obtained in the previous section to calculate the photoexcitation of a multielectron atom by an ultrashort Gaussian pulse of radiation (2.30) in the local plasma frequency model. Within the framework of this model the expression for the photoabsorption cross-section of the atom is [3]

$$\sigma_{ph}^{(BL)}(\omega') = \frac{2\pi^2 e^2}{mc} \int n(r) \delta(\omega' - \omega_{pl}(r)) dr, \quad (2.43)$$

where $\omega_{pl}(r) = \sqrt{4\pi e^2 n(r)/m}$ is the local plasma frequency and $n(r)$ is the spatial distribution of electron density in the atom. Substituting (2.43) into (2.19), we find

$$W_{tot}^{(ph)} = \frac{\sqrt{\pi} e}{\sqrt{m} \hbar} \int_0^\infty |E(\omega_{pl}(r), \varphi)|^2 \sqrt{n(r)} r^2 dr, \quad (2.44)$$

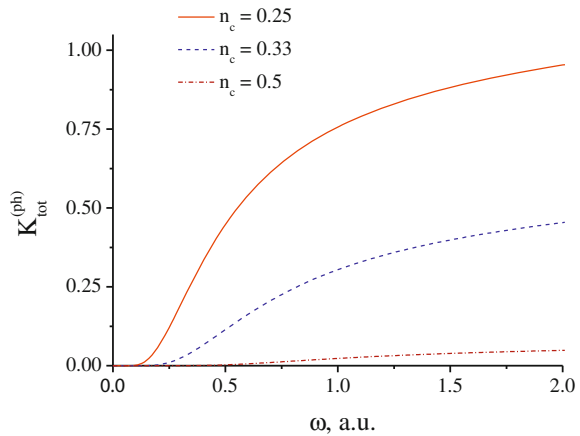
where $|E(\omega_{pl}(r), \varphi)|^2$ is the squared magnitude of the Fourier transform of the electric field in a pulse of the form (2.31), calculated at the local plasma frequency, in which the CE phase dependence is explicitly specified. To analyze the phase effects in the total probability of photoexcitation by ultrashort laser pulses, we introduce the phase modulation factor by the formula

$$K_{tot}^{(ph)} = 2 \frac{W_{tot}^{(ph)}(\varphi = 0) - W_{tot}^{(ph)}(\varphi = \pi/2)}{W_{tot}^{(ph)}(\varphi = 0) + W_{tot}^{(ph)}(\varphi = \pi/2)}. \quad (2.45)$$

The phase modulation factor of the total probability of photoabsorption by an atom with charge $Z = 30$ calculated using the statistical model for the electron density is presented in Fig. 2.1 for three pulse widths as a function of the carrier frequency. It will be recalled that the dimensionless parameter n_c is the number of periods at the carrier frequency in the radiation pulse. We note that appreciable dependence of the photoabsorption probability on the CE phase occurs only for $n_c < 0.5$, and also in the case of a bound-bound transition. The phase modulation factor for the fixed parameter n_c grows with carrier frequency. It should be noted that the probability of photoabsorption at the high-frequency boundary of the interval presented in Fig. 2.1 is 15 % of its maximum value, corresponding in this model to the frequency $\omega_{\max} = 0.4$ a.u.

The expression (2.19) for the total photoabsorption probability can be applied to the interaction of an ultrashort pulse with a *metal nanosphere* in a dielectric medium. When the radiation wavelength is much longer than the nanoparticle radius r_s , its dynamic polarizability can be described by the Lorentz formula

Fig. 2.1 Phase modulation factor for the total probability of photoabsorption of an ultrashort pulse by an atom as a function of the carrier frequency



$$\beta_s(\omega) = \varepsilon_m \frac{\varepsilon_s(\omega) - \varepsilon_m}{\varepsilon_s(\omega) + 2\varepsilon_m} r_s^3, \quad (2.46)$$

where $\varepsilon_s(\omega)$ is the dielectric permittivity of the nanoparticle metal and ε_m is the dielectric permittivity of the matrix. Hence we can use the optical theorem to find the photoabsorption cross-section in the dipole approximation and then (2.19) to calculate the total photoabsorption probability for the whole time of action of the pulse.

The results of calculations to find the probability of photoabsorption of an ultrashort pulse by a silver nanoparticle in a glass matrix are shown in Fig. 2.2 for two values of the CE phase. The frequency dependence of the dielectric permittivity of silver is restored using data for the real and imaginary parts of the refractive index.

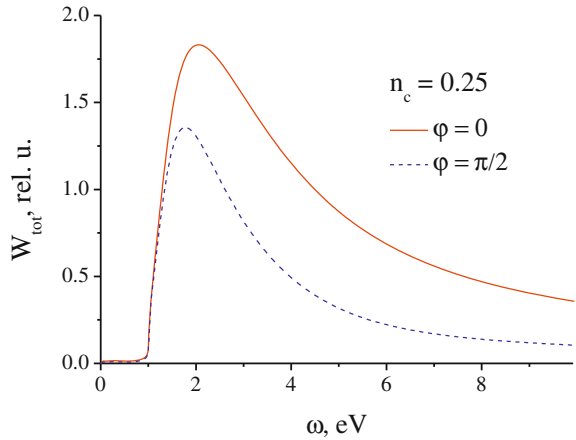
We thus that in this case ($n_c = 0.25$) photoabsorption depends heavily on the CE phase, especially for photon energies at carrier frequencies exceeding the maximum energy. With increasing pulse duration, the phase dependence of the probability becomes less noticeable, and for $n_c > 0.5$ it practically disappears.

In a number of cases quantum systems are excited by a sequence of identical pulses separated by some time interval T (not to be confused with the oscillation period). It is not difficult to obtain a Fourier transform of the electric field strength for such a sequence consisting of N identical pulses in terms of the Fourier transform of a single pulse $E(\omega')$:

$$E_N(\omega') = \frac{\sin(\omega' T N/2)}{\sin(\omega' T/2)} \exp\left[i \frac{(N-1)\omega' T}{2}\right] E(\omega'). \quad (2.47)$$

Substituting (2.47) into the right-hand side of (2.19), we find the probability of photoexcitation of a quantum transition under the action of N identical pulses:

Fig. 2.2 Total probability of photoabsorption of an ultrashort pulse ($n_c = 0.25$) on a silver sphere ($r_s = 5.3$ nm) as a function of the carrier frequency for two values of the CE phase



$$W_{21}(N) = \frac{c}{4\pi^2 \hbar} \int \frac{\sigma_{21}(\omega')}{\omega'} \left[\frac{\sin(\omega' T N/2)}{\sin(\omega' T/2)} \right]^2 |E(\omega')|^2 d\omega'. \quad (2.48)$$

We now use these expressions to describe photoionization of a hydrogen atom under the action of a series of short pulses. In this case the process cross-section $\sigma_{21}(\omega)$ is given by the Sommerfeld formula

$$\sigma_H\left(v = \frac{\hbar \omega}{Ry}\right) = \frac{2^9 \pi^2 a_B^2}{3 \cdot 137 \cdot v^4} \frac{\exp\left(-\frac{4 \arctg \sqrt{v-1}}{\sqrt{v-1}}\right)}{1 - \exp(-2\pi/\sqrt{v-1})}, \quad (2.49)$$

where $a_B \cong 0.53 \text{ \AA}$ is the Bohr radius and $Ry \cong 13.6 \text{ eV}$ denotes the Rydberg energy. Figure 2.3 shows the results of calculations using (2.48) and (2.49) to find the probability of photoionization of a hydrogen atom by a series of laser pulses with a width of two oscillation periods at the carrier frequency. Plotted on the abscissa is the value $v = \varepsilon/Ry + 1$, where $\varepsilon = \hbar \omega - Ry$ is the energy of an ionized electron. Clearly, with a growing number of pulses, the spectral dependence of the photoexcitation probability narrows near the maximum value determined from the equation $\omega T = 2\pi k$ (where k is a natural number). Since the energy of an electron knocked out of an atom is $Ry(v - 1)$ and the number of these electrons is proportional to the probability $W(N)$, the figure shows that the energy spectrum of the photoelectrons can be controlled by changing the parameters of the series of exciting pulses.

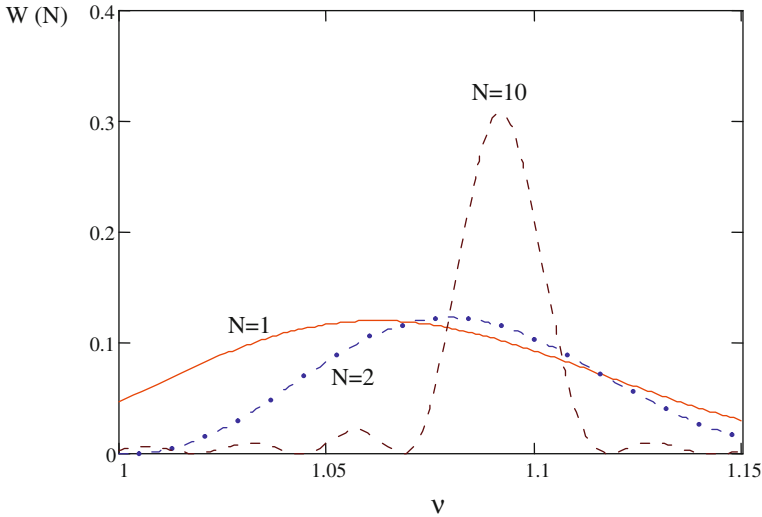


Fig. 2.3 Probability of photoionization of a hydrogen atom under the action of a sequence of N two-cycle laser pulses, $v = \hbar \omega / Ry$

2.3 Dependence of the Excitation Probability on the Duration of Ultrashort Pulses

We now consider the dependence of the photoexcitation probability on the duration of ultrashort Gaussian pulses (2.30) for the example of a multielectron atom in the Lenz–Jensen statistical model [4]. For the photoabsorption cross-section we use the local plasma frequency model (2.43). Then the process probability is determined by the basic formula (2.19) and the Fourier transform of the electric field strength in the pulse.

2.3.1 Atom in the Lenz–Jensen Statistical Model

Common to all statistical models of an atom, the expression for the local electron density has the form

$$n_{LJ}(r) = Z^2 f(x = r/r_{TF}) a_B^{-3}, \quad (2.50)$$

$$r_{TF} = \frac{b}{\sqrt[3]{Z}} a_B, \quad b = \sqrt[3]{\frac{9\pi^2}{128}} \cong 0.8853$$

where Z is the charge on the atomic nucleus, r_{TF} is the Thomas–Fermi radius, $a_B = \frac{\hbar^2}{me^2}$ is the Bohr radius, and $f(x)$ is an universal function of the dimensionless distance to the nucleus $x = r/r_{TF}$ depending on the specific statistical model of the atom. Lenz and Jensen proposed the following expression for this function:

$$f_{LJ}(x) \cong 3.7e^{-\sqrt{9.7x}} \frac{(1 + 0.26\sqrt{9.7x})^3}{(9.7x)^{3/2}}. \quad (2.51)$$

This provides a more realistic description of the electron density distribution in a multielectron atom than the Thomas–Fermi function. The spectral photoabsorption cross-section of an atom in the Lenz–Jensen model calculated using (2.43), (2.50), and (2.51) is presented in Fig. 2.4 for two values of the charge on the atomic nucleus.

2.3.2 Total Photoabsorption Cross-Section

We now write down the expression for the total photoabsorption cross-section (2.19) separating out the dependence on the duration of the ultrashort pulses as expressed through the number of cycles n_c [see the determination of the number of cycles (2.34)]:

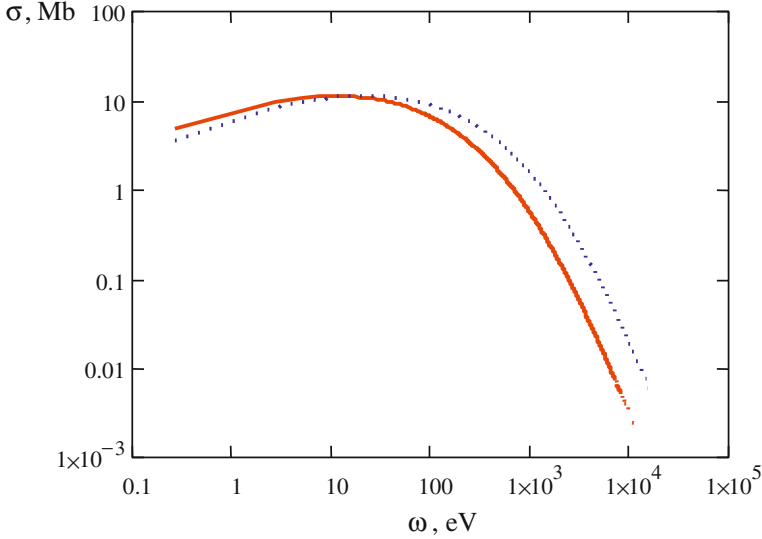


Fig. 2.4 Spectral photoabsorption cross-section of a Lenz-Jensen atom calculated in the local plasma frequency model for two nuclear charges: *solid line* $Z = 30$, *dotted line* $Z = 60$

$$W_{tot}(n_c) = \frac{c}{(2\pi)^2} \int_0^\infty \sigma(\omega) \frac{|\mathbf{E}(\omega, n_c)|^2}{\hbar \omega} d\omega, \quad (2.52)$$

where $\sigma(\omega)$ is the spectral cross-section of the process, c is the velocity of light, and $\mathbf{E}(\omega, n_c)$ is the Fourier transform of the electric field strength in the pulse.

The advantage of ultrashort pulse width measurements in terms of the number of pulses in the expression for the total photoabsorption probability consists in the universal nature of the resulting dependences, which hold over a wide spectral range.

Once normalized (to the squared magnitude of the Fourier transform of the electric field strength peak value $|\mathbf{E}_0(\omega)|^2$), the total photoprocess probability in the monochromatic limit $n_c \gg 1$ can be written

$$\tilde{W}_{tot}^{(mon)} = \frac{W_{tot}^{(mon)}}{|\mathbf{E}_0(\omega)|^2} \rightarrow \frac{c \sigma(\omega)}{\hbar \omega^2} n_c. \quad (2.53)$$

Thus the photoprocess probability in the monochromatic limit (the long pulse limit) grows linearly with the number of cycles n_c in the pulse, as expected from traditional considerations. Moreover, in the monochromatic limit the photoprocess probability does not depend on the CE phase φ .

In the opposite limiting case of a pulse of zero width ($n_c \ll 1$) and constant CE phase ($\Phi = \varphi = \text{const}$), it can be shown that (2.36)–(2.38) lead to

$$W_{tot}(n_c) = \frac{4\pi}{3} \left(\frac{E_0}{\hbar\omega} \right)^2 n_c^2 \langle 0 | \hat{\mathbf{d}}^2 | 0 \rangle \cos^2 \varphi, \quad (2.54)$$

where $\langle 0 | \hat{\mathbf{d}}^2 | 0 \rangle$ is the quantum-mechanical mean value of the squared magnitude of the atomic electron electric dipole moment operator and E_0 is the electric field amplitude in the pulse. The sum rule was used to derive the right-hand side of (2.54).

Thus in the limit of a pulse of zero width the total photoprocess probability is proportional to the *squared* number of cycles in the pulse and the squared cosine of the CE phase.

Here we are interested in the dependence of the total photoprocess probability on the parameter n_c in the low value region $n_c \sim 1$ and $n_c \leq 1$. To determine this dependence, we use the above formulas describing photoabsorption in the framework of the local plasma frequency model, and with the atomic electron density in the Lenz–Jensen model. The results of our calculations are given in Figs. 2.5, 2.6, 2.7, and 2.8 for a Lenz–Jensen atom with nuclear charge $Z = 30$ and normalized total photoabsorption probability \tilde{W}_{tot} determined by

$$\tilde{W}_{tot} = \frac{W_{tot}}{|\mathbf{E}_0|^2}. \quad (2.55)$$

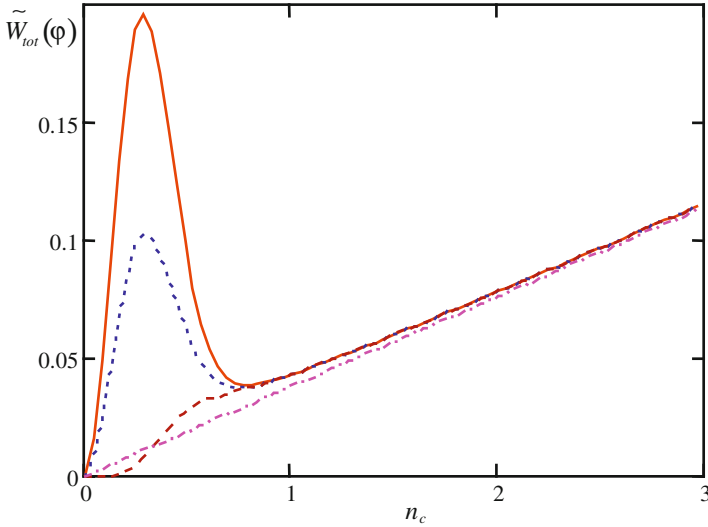


Fig. 2.5 Total photoabsorption probability for ultrashort pulses with different values of the CE phase, calculated for a Lenz–Jensen atom: *solid line* $\varphi = 0$, *dotted line* $\varphi = \pi/4$, *dashed line* $\varphi = \pi/2$, *dash-and-dot line*—monochromatic limit, $\hbar\omega = 10$ a.u., $Z = 30$

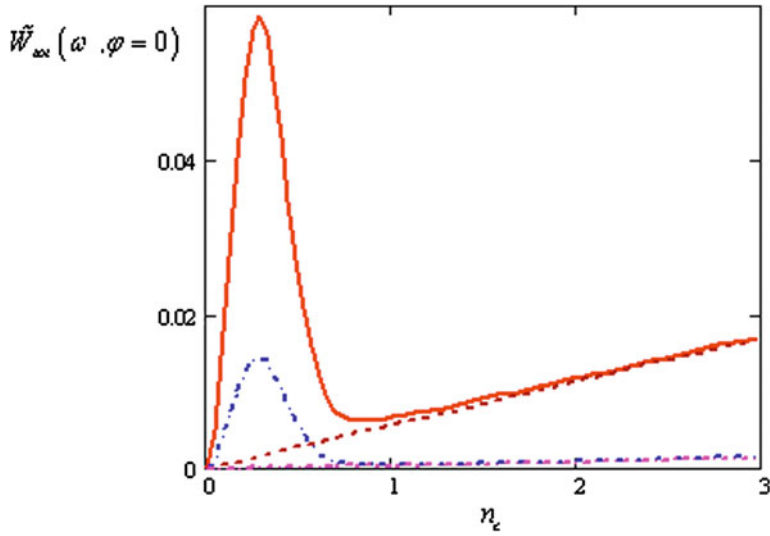


Fig. 2.6 Total photoabsorption probability for ultrashort pulses with the CE phase equal to zero, at two different carrier frequencies and for a Lenz–Jensen atom: *solid line* $\hbar\omega = 500$ eV, *dash-and-dot line* $\hbar\omega = 1000$ eV. *Straight lines* show corresponding monochromatic limits

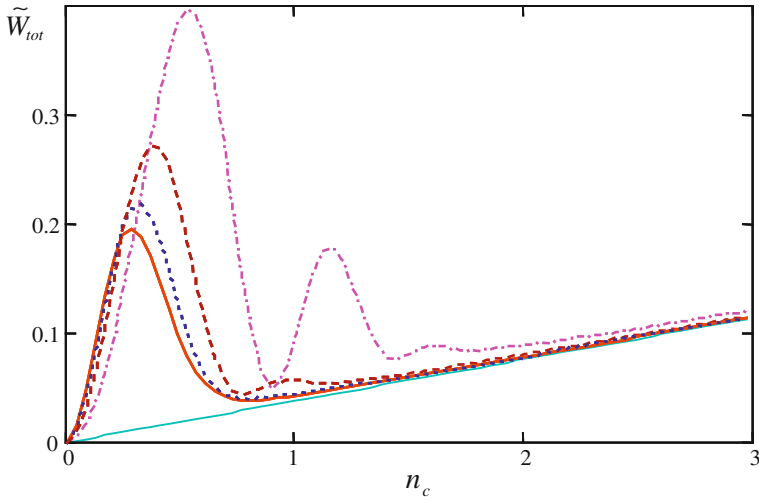


Fig. 2.7 Total photoabsorption probability for chirped ultrashort pulses and for a Lenz–Jensen atom, $\hbar\omega = 10$ a.u., $Z = 30$: *solid red line* $\alpha = 0$, *dotted line* $\alpha = 0.5$, *dashed line* $\alpha = 1$, *dash-and-dot line* $\alpha = 2$, *solid cyan line*—monochromatic limit

The probability of photoabsorption under the action of ultrashort pulses with constant CE phase $\Phi(t) = \varphi = \text{const}$ is shown in Fig. 2.5 for a specified value of the carrier frequency and different values of the parameter φ .

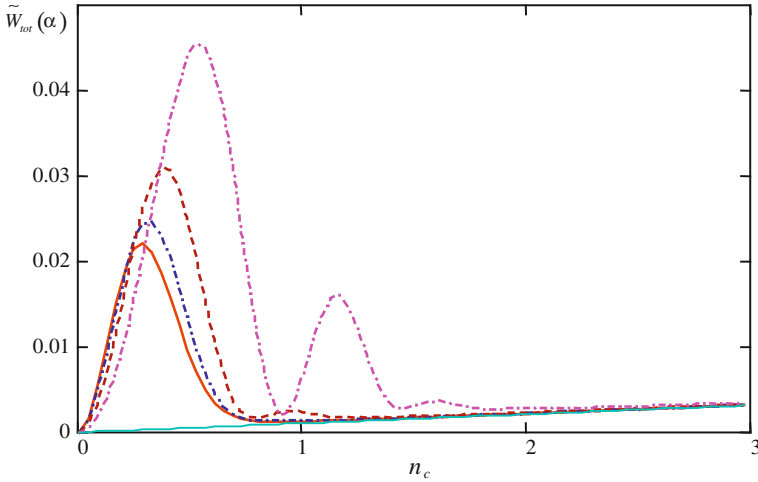


Fig. 2.8 Total photoabsorption probability for chirped ultrashort pulses and for a Lenz–Jensen atom, $\hbar\omega = 30$ a.u., $Z = 30$: solid red line $\alpha = 0$, dotted line $\alpha = 0.5$, dashed line $\alpha = 1$, dash-and-dot line $\alpha = 2$, solid cyan line—monochromatic limit

For subcycle pulses, it can be seen that the probability depends strongly on the CE phase. For example, for a cosine pulse ($\varphi = 0$), photoabsorption has a maximum at $n_c \leq 0.5$. The amplitude of this maximum decreases with growing CE phase.

The straight line in Figs. 2.5, 2.6, 2.7, and 2.8 represents the photoabsorption probability in the monochromatic limit, when the function $\tilde{W}(n_c)$ has linear behavior [see (2.53)]. As can be seen from Fig. 2.5, the total photoabsorption probability tends to a straight line for $n_c \geq 1$, as it should in the monochromatic limit. Thus in the case considered, the probability $\tilde{W}(n_c)$ depends nonlinearly on the pulse width (the parameter n_c) only for subcycle ultrashort pulses.

Figure 2.6 shows the dependence of the total probability of atomic photoabsorption on the parameter n_c for a cosine pulse ($\varphi = 0$) and different carrier frequencies ω expressed in electron-volts. From this figure it follows in particular that, for the case under consideration, the function $\tilde{W}(n_c)$ has a smaller angle of inclination to the X axis for higher values of the carrier frequency.

2.3.3 Excitation of an Atom by a Chirped Pulse

Let us consider the dependence of the photoexcitation probability on the duration of ultrashort pulses when an atom is acted on by a chirped electromagnetic pulse and the initial phase Φ is quadratically time-dependent: $\Phi(t) = kt^2$. The Fourier transform of the electric field strength in such a pulse is given by (1.34).

The results of the corresponding calculations are shown in Figs. 2.7 and 2.8 for different values of the carrier frequency in the Lenz–Jensen model.

We see that the function $\tilde{W}(n_c)$ is in this case somewhat more complex than for a fixed CE phase (compare with Figs. 2.5 and 2.6). For example, for high enough values of the dimensionless chirp α there are several maxima of the photoabsorption probability as a function of the number of cycles in a pulse, and the value of these maxima decreases with growing parameter n_c . It is significant that for a chirped pulse the photoabsorption probability maxima occur not only for subcycle ultrashort pulses, but also in the region $n_c > 1$.

In the case of a multicycle chirped pulse, coincidence of the process probability with the monochromatic limit is retained for high enough values of the parameter n_c . However, the number of cycles in a pulse at which this coincidence begins to occur depends on the value of the dimensionless chirp $\alpha = \kappa \Delta t^2$ (see (1.34)). The higher the chirp value, the more cycles are required in the pulse to reach the monochromatic limit (linear dependence of the probability $\tilde{W}(n_c)$ on the parameter n_c).

From comparison of Figs. 2.7 and 2.8 it follows that, as for the constant CE phase, in the monochromatic limit the photoabsorption dependence on the number of cycles in a pulse becomes weaker with growing carrier frequency. This is explained by reduction of the photoabsorption cross-section of a Lenz–Jensen atom with growing frequency in the spectral range $\omega > 10$ eV (see Fig. 2.5).

2.4 Scattering of Ultrashort Pulses by Atoms and in a Plasma

Most works treating the interaction of ultrashort pulses with a material substance are devoted to photoionization and photoexcitation of atomic particles [1]. The description of photoprocesses within the framework of the perturbation theory then has a rather limited domain of applicability since, for characteristic values of the radiation intensity used in modern experiments, nonlinear effects are rather significant.

For radiation scattering by atomic particles, due to the low value of the process cross-section, the perturbation theory turns out to be applicable over a much wider region of parametric variation than in photoionization and photoexcitation. However, application of the usual formulas of perturbation theory obtained in the long pulse limit in the case of single-cycle and subcycle pulses becomes, generally speaking, incorrect. In this section we develop a method to describe scattering of ultrashort electromagnetic pulses by an atom and in a plasma, taking into account possible excitation of the target and the non-dipole nature of the electromagnetic interaction.

2.4.1 Scattering of an Ultrashort Pulse by an Atom

Let us consider scattering of an ultrashort electromagnetic pulse by an atom, taking into account possible excitation of the target [5]. We assume that the spatio-temporal dependence of the electric field strength in the pulse has the form

$$\mathbf{E}(t, \mathbf{r}) = \mathbf{e} E_0 g\left(t - \frac{\mathbf{n}\mathbf{r}}{c}\right), \quad (2.56)$$

where \mathbf{e} is the unit polarization vector, E_0 is the field amplitude, \mathbf{n} is the unit vector in the direction of the electromagnetic pulse propagation, $g(\tau)$ is the dimensionless function defined by a concrete realization of the pulse, and c is the velocity of light.

We will decompose the strength (2.56) into plane waves with frequencies ω and wave vectors $\mathbf{k} = (\omega/c)\mathbf{n}$. Then scattering of an electromagnetic field pulse can be represented as scattering of a set of plane waves to a plane wave with frequency ω' , unit polarization vector \mathbf{e}' , and wave vector $\mathbf{k}' = (\omega'/c)\mathbf{n}'$.

With the above picture, the differential probability of scattering for the whole time of action of the pulse with simultaneous excitation of the target from the state $|i\rangle$ to the state $|f\rangle$ can be obtained in the form

$$\frac{dW_{fi}}{d\Omega' d\omega'} = \int_0^\infty \frac{d\sigma_{fi}(\mathbf{k}', \mathbf{k})}{d\Omega' d\omega'} \frac{dN_{ph}}{d\omega dS} d\omega, \quad (2.57)$$

where

$$\frac{d\sigma_{fi}(\mathbf{k}', \mathbf{k})}{d\Omega' d\omega'} = \delta(\omega - \omega' - \omega_{fi}) \frac{\omega'^3 \omega}{c^4} \left| e_l'^* e_s c_{fi}^{ls}(\mathbf{k}', \mathbf{k}) \right|^2 \quad (2.58)$$

is the differential scattering cross-section with respect to the solid angle and frequency for a plane wave, with $c_{fi}^{ls}(\mathbf{k}', \mathbf{k})$ the radiation scattering tensor taking into account excitation of the target, and

$$\frac{dN_{ph}}{d\omega dS} = \frac{c}{(2\pi)^2} \frac{|\mathbf{E}(\omega)|^2}{\hbar \omega} \quad (2.59)$$

is the number of photons forming the electromagnetic pulse field in the spectral range $(\omega, \omega + d\omega)$ that passed through the unit area during the whole time of action of the radiation, with $\mathbf{E}(\omega)$ the Fourier transform of the electric field strength. Substituting (2.58) into the right-hand side of (2.57), we arrive at an expression for the differential photoprocess probability over the whole time of action of the field that generalizes the expression obtained in [6] to take into account excitation of the target and the non-dipole nature of the electromagnetic interaction.

Gathering the results (2.56)–(2.58), we find the following basic equation:

$$\frac{dW_{fi}}{d\Omega' d\omega'} = \frac{\omega'^3}{c^3} \frac{E_0^2}{4\pi^2 \hbar} |g(\omega' + \omega_{fi})|^2 |e_l'^* e_s c_{fi}^{ls}(\mathbf{k}', \mathbf{k})|^2, \mathbf{k} = \frac{\omega' + \omega_{fi}}{c} \mathbf{n}, \quad (2.60)$$

where $g(\omega)$ is the Fourier transform of the temporal pulse shape function $g(\tau)$ and twice-repeated indices are summed over.

Equation (2.60) can also be obtained using second order perturbation theory in a consistent quantum–mechanical approach [6].

Hereafter we assume that the scattering tensor reduces to the scalar $c_{fi}^{ls} = \delta_{ls} c_{fi}$ ($c_{fi} = (1/3) c_{fi}^{ll}$), with summation over twice-repeated indices. Then on the right-hand side of (2.60) we have a scalar product of polarization vectors of incident and scattered waves, averaged in the usual way for non-polarized scattered radiation.

We consider two cases: (1) scattering without excitation of the target (so-called elastic scattering) and (2) scattering with excitation of the target into an arbitrary state.

For scattering without change of the atomic state and in the multiplicative approximation, we have

$$c_{ii}(\mathbf{k}', \mathbf{k}) \simeq \beta_i(\omega') \tilde{F}_{ii}(\mathbf{k}' - \mathbf{k}), \quad (2.61)$$

where $\beta_i(\omega')$ is the dynamic dipole polarizability of an atom in the initial state and $\tilde{F}_{ii}(\mathbf{q}) = F_{ii}(\mathbf{q})/Z$ is the atomic form factor normalized by the number of electrons. In view of (2.61), after summation over polarizations of the scattered photon, (2.60) implies

$$\frac{dW_{ii}}{d\Omega' d\omega'} = \frac{1 - (\mathbf{e}\mathbf{n}')^2}{4\pi^2} \left(\frac{\omega'}{c}\right)^3 \frac{E_0^2}{\hbar} |g(\omega')|^2 |\beta_i(\omega')|^2 \tilde{F}_{ii}^2 \left(2 \frac{\omega'}{c} \sin\left(\frac{\theta}{2}\right)\right), \quad (2.62)$$

where θ is the scattering angle. Here, when we write Δk in the argument of the atomic form factor, we take into account the fact that $\omega' = \omega$.

The formula (2.62) can be rewritten in terms of the atomic polarization charge

$$Z_{pol}(\omega) = \frac{m\omega^2}{e^2} |\beta(\omega)| \quad (2.63)$$

as

$$\frac{dW_{ii}}{d\Omega' d\omega'} = \frac{1 - (\mathbf{e}\mathbf{n}')^2}{4\pi^2} \left(\frac{e^2}{\hbar c}\right)^3 \left(\frac{I_0}{I_a}\right) Z_{pol}^2(\omega') \frac{|\omega_a g(\omega')|^2}{\omega'} \tilde{F}_{ii}^2 \left(2 \frac{\omega'}{c} \sin\left(\frac{\theta}{2}\right)\right), \quad (2.64)$$

where $I_a = c \frac{m^4 e^{10}}{8\pi \hbar^8} \simeq 3.5 \cdot 10^{16} \text{ W/cm}^2$, $\omega_a = \frac{m e^4}{\hbar^3}$ are the atomic units of radiation intensity and frequency, and $I_0 = c E_0^2 / 8\pi$ is the mean radiation intensity. In

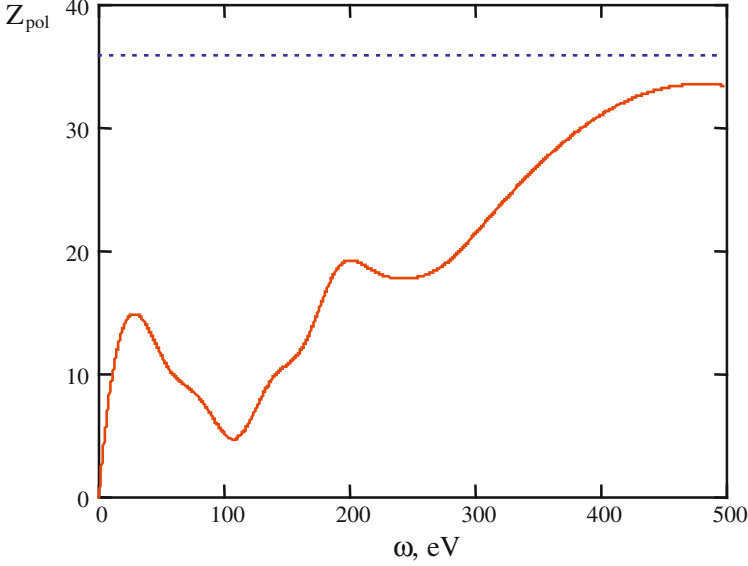


Fig. 2.9 Polarization charge of a krypton atom as a function of frequency. The *dashed line* shows the number of electrons in the atom

the high-frequency limit $\omega \gg \omega_t$ (ω_t is the characteristic eigenfrequency of the target), the polarization charge is equal to the number of electrons in the target.

The frequency dependence of the polarization charge of a krypton atom calculated using experimental photoabsorption data is presented in Fig. 2.9. The solid curve in the figure is obtained by calculating the imaginary part of the polarizability of the krypton atom with the optical theorem and restoring the real part by means of the Kramers–Kronig relation.

When the carrier frequency ω_c of the pulse is close to one of the eigenfrequencies of excitation of the atom in the discrete spectrum $\omega_c \approx \omega_{ri}$ (in this case the oscillator strength for the corresponding transition is nonzero $f_{ri} \neq 0$), the resonant approximation can be used for the polarizability:

$$|\beta_i(\omega' \approx \omega_{ri})|^2 \cong \frac{\pi}{2} \left(\frac{e^2}{m \omega'^2} \right)^2 \frac{\omega'}{\delta_{ri}} f_{ri}^2 \omega' G_{ri}(\omega'), \quad (2.65)$$

where δ_{ri} and $G_{ri}(\omega)$ are the width and shape of the resonance transition line. Then instead of (2.62) we have

$$\frac{dW_{ii}(\omega_c \approx \omega_{ri})}{d\Omega' d\omega'} = \frac{1 - (\mathbf{e} \mathbf{n}')^2}{8 \pi} \left(\frac{e^2}{\hbar c} \right)^3 \left(\frac{I_0}{I_a} \right) \frac{\omega'}{\delta_{ri}} |\omega_a g(\omega')|^2 f_{ri}^2 G_{ri}(\omega'). \quad (2.66)$$

In writing (2.66), we have taken into account the fact that, in the spectral range under consideration, the atomic form factor can be assumed equal to unity.

In particular, the resulting formula implies resonant amplification of scattering due to the presence of the multiplier ω'/δ_{ri} .

For an ultrashort pulse with carrier frequency in the optical range, the spectrum is generally much broader than the line of a resonance transition in an atom, so the spectral dependence of the scattering probability will be defined mainly by the shape $G_{ri}(\omega')$ of the spectral line of the transition. In the general case, the spectrum of a scattered pulse will also be influenced by the function $|g(\omega')|^2$.

The resonance probability (2.66) can be generalized to the case where the atom is excited in the scattering process if the frequency of scattered radiation is close to one of the eigenfrequencies for transition of the atom from the intermediate state to the final state.

The probability of scattering with target excitation to an arbitrary state (the entire scattering spectrum) is obtained by summing the probability (2.60) over all possible states $|f\rangle$. Let us consider the entire scattering spectrum in the high-frequency range ($\omega \gg \omega_i$), when the approximate expression for the electromagnetic field scattering tensor is valid:

$$c_{fi}^{(hf)}(\mathbf{k}', \mathbf{k}) \simeq -\frac{e^2}{m \omega' \omega} F_{fi}(\mathbf{k}' - \mathbf{k}). \quad (2.67)$$

Substituting the right-hand side of (2.67) into the formula (2.60) and summing over all possible final states, we find the following expression for the scattering probability with excitation of the atom:

$$\frac{dW_{tot}^{(hf)}}{d\omega' d\Omega'} = \frac{c E_0^2}{4\pi^2 \hbar} \left[1 - (\mathbf{e} \mathbf{n}')^2 \right] r_e^2 \int_0^\infty |g(\omega)|^2 S_i(\omega' - \omega, \mathbf{k}' - \mathbf{k}) \frac{d\omega}{\omega}, \quad (2.68)$$

where

$$S_i(\Delta\omega, \Delta\mathbf{k}) = \frac{1}{2\pi} \int_{-\infty}^\infty dt e^{-i\Delta\omega t} \langle i | \hat{n}(\Delta\mathbf{k}, t) \hat{n}(-\Delta\mathbf{k}) | i \rangle \quad (2.69)$$

is the dynamic form factor (DFF) of an atom in the i th state (see Appendix III).

In the elementary approximation, the DFF of a hydrogen-like atom is

$$S(\Delta\omega, \Delta\mathbf{k}) \simeq \delta\left(\Delta\omega + \frac{\hbar}{2m} \Delta\mathbf{k}^2\right). \quad (2.70)$$

Substituting the right-hand side of (2.70) into (2.68) and integrating with respect to the frequency ω , we find

$$\frac{dW_{tot}^{(hf)}}{d\omega' d\Omega'} \approx \frac{2}{\pi} \left[1 - (\mathbf{e} \mathbf{n}')^2 \right] \frac{I_0}{\hbar} r_e^2 \frac{|g[\omega(\omega', \theta)]|^2}{\omega(\omega', \theta)}, \quad (2.71)$$

where

$$\omega(\omega', \theta) = \omega_r + \omega' \cos \theta - \omega_r \sqrt{1 - 4 \frac{\omega'}{\omega_r} \sin^2\left(\frac{\theta}{2}\right) - \left(\frac{\omega'}{\omega_r}\right)^2 \sin^2(\theta)},$$

$$\omega_r = \frac{m c^2}{\hbar} \quad (2.72)$$

For $\omega' \ll \omega_r$, we have $\omega(\omega', \theta) \cong \omega'$, and instead of (2.71) we obtain

$$\frac{dW_{tot}^{(hf)}}{d\omega' d\Omega} \approx \frac{1 - (\mathbf{e}\mathbf{n}')^2}{4\pi^2} \left(\frac{e^2}{\hbar c}\right)^3 \left(\frac{I_0}{I_a}\right) \frac{|\omega_a g(\omega')|^2}{\omega'}. \quad (2.73)$$

This expression describes the entire spectrum for scattering of an ultrashort pulse by a one-electron atom in the high-frequency approximation.

In the high-frequency limit, when $Z_{pol} \cong Z$, the ratio of the probability of “elastic” scattering of an ultrashort pulse by a hydrogen-like atom (2.64) to the total probability (2.73) is equal to the squared normalized form factor of the atom:

$$R^{(hf)}(\omega', \theta) \equiv \frac{dW_{ii}^{(hf)}}{dW_{tot}^{(hf)}} = \tilde{F}_{ii}^2 \left(2 \frac{\omega'}{c} \sin\left(\frac{\theta}{2}\right) \right). \quad (2.74)$$

Let us consider scattering of an ultrashort Gaussian electromagnetic pulse by an atom. Then the function determining the time dependence of the electric field strength in (2.56) looks like

$$g(\tau) = \exp(-\tau^2/\Delta t^2) \cos(\omega_c \tau + \varphi), \quad (2.75)$$

where ω_c is the carrier frequency, Δt is the time parameter proportional to the pulse width, and φ is the carrier phase with respect to the envelope (the CE phase). It is convenient to express the parameter Δt in terms of the number of periods in the pulse at the carrier frequency n_c : $\Delta t = 2\sqrt{2}\pi n_c/\omega_c$. In view of this fact, the squared magnitude of the Fourier transform of the function (2.75) appearing in the expressions for the scattering probability can be represented as

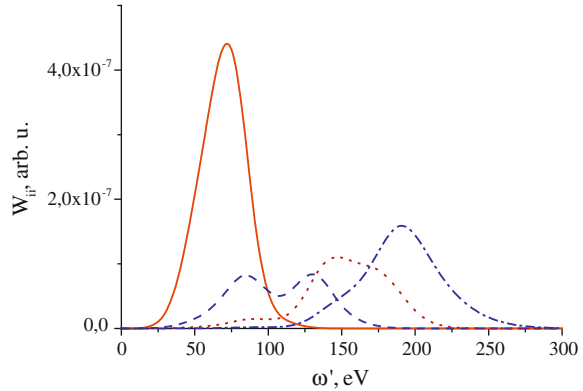
$$|g(\omega)|^2 = 2\pi^2 \left(\frac{n_c}{\omega_c}\right)^2 G_E(\omega, \omega_c, n_c) [1 + K_{ph}(\omega, \omega_c, n_c) \cos(2\varphi)], \quad (2.76)$$

where

$$G_E(\omega, \omega_c, n_c) = \exp\left[-4\pi n_c^2 \left(1 - \frac{\omega}{\omega_c}\right)^2\right] + \exp\left[-4\pi n_c^2 \left(1 + \frac{\omega}{\omega_c}\right)^2\right] \quad (2.77)$$

is the spectral form of the pulse and

Fig. 2.10 Spectrum of “elastic” scattering of a single-cycle pulse by a krypton atom for different values of the carrier frequency: *solid curve* $\hbar\omega_c = 80$ eV, *dashed curve* $\hbar\omega_c = 110$ eV, *dotted curve* $\hbar\omega_c = 140$ eV, *dash-and-dot curve* $\hbar\omega_c = 180$ eV



$$K_{ph}(\omega, \omega_c, n_c) = \text{sech}\left(8\pi n_c^2 \frac{\omega}{\omega_c}\right) \quad (2.78)$$

is effectively the phase modulation factor. It follows from (2.78) that the phase modulation factor has an appreciable value only for ultrashort pulses, when $n_c \sim 1$.

Figure 2.10 presents the spectrum of “elastic” scattering (i.e., without target excitation) of a single-cycle pulse by a krypton atom, as calculated using (2.62) for a scattering angle of 45° and several values of the carrier frequency.

As can be seen from this figure, in the case of a single-cycle pulse, the form of the scattered radiation spectrum essentially depends on the carrier frequency value. Far from the minimum of the frequency dependence of the polarization charge of a krypton atom, which falls approximately on 107 eV (see Fig. 2.9), the spectral scattering curves have a symmetric form with a maximum at the centre. Near the minimum frequency ($\hbar\omega_c = 110$ eV), a dip appears in the scattering spectrum. It follows from (2.64) that the described evolution of the scattering spectrum is explained by superposition of two frequency dependences: one is the ultrashort pulse spectrum (2.76) and the other the atomic polarization charge spectrum. For a single-cycle pulse with spectral width broad enough to be comparable with the scale of spectral singularities of the krypton atom polarization charge, this superposition modifies the form of the spectral scattering curve. The situation changes in going to longer pulses, for example, to a three-cycle pulse, whose spectrum for scattering by a krypton atom is presented in Fig. 2.11 for different carrier frequencies.

We see that for a three-cycle pulse the scattered radiation spectrum is a bell-shaped curve. Its shape is defined by the spectrum of the incident pulse (2.76) and the amplitude depends on the value of the polarization charge of the atom at the carrier frequency.

Figure 2.12 shows the result of calculations using the expression (2.74) for the ratio of the probability of “elastic” scattering of a single-cycle pulse by a hydrogen atom in the high-frequency limit summed over all scattered radiation frequencies to the analogous value for the process with arbitrary excitation of the atom.

Fig. 2.11 The same as Fig. 2.10 but for a three-cycle pulse

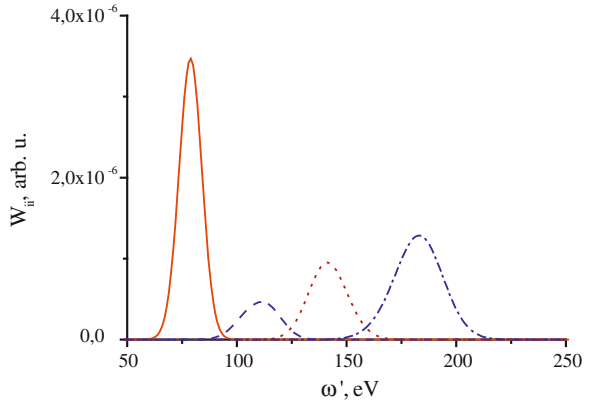
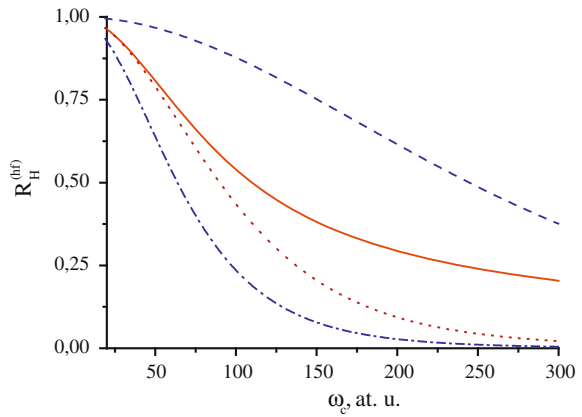


Fig. 2.12 Ratio of the probability of “elastic” scattering of a single-cycle pulse by a hydrogen atom to the total probability calculated in the high-frequency limit for three scattering angles and for probabilities integrated with respect to the angles (*solid curve*): *dashed curve* $\theta = 30^\circ$, *dotted curve* $\theta = 90^\circ$, *dash-and-dot curve* $\theta = 180^\circ$



This ratio is calculated as a function of the carrier frequency of the pulse for different scattering angles. Shown in the same figure by a solid curve is the contribution of the “elastic” process summed over the scattering angles. From Fig. 2.12 we see that, for low values of the carrier frequency of the electromagnetic pulse, scattering occurs mainly without excitation of the atom. For wide scattering angles, the role of the elastic channel decreases more rapidly with growing frequency. For the process probability integrated with respect to the angles, the contributions of the elastic and inelastic channels are compared for a carrier frequency of about 112 a.u., which corresponds to a photon energy of about 3 keV.

2.4.2 Scattering of an Ultrashort Pulse in a Plasma

Here we use the approach developed for an atomic target to describe scattering of an ultrashort pulse in a plasma [7]. We proceed from the formula (2.68), obtained

in the high-frequency approximation for an atom. It will be recalled that, since plasma electrons are free, the high-frequency condition $\omega \gg \omega_t = 0$ (ω_t is the characteristic eigenfrequency of a plasma electron) is satisfied automatically for them, so (2.68) is applicable. In this expression the dynamic form factor of an atom should be replaced by the dynamic form factor of an electron component of the plasma. This is given by (A.3) and (A.19) in Appendix III.

Scattering of electromagnetic radiation in a plasma can be of two types: Compton scattering and transition scattering. Compton scattering corresponds to large variations of the wave vector $|\Delta \mathbf{k}| > r_D^{-1}$ ($\Delta \mathbf{k} = \mathbf{k}' - \mathbf{k}$), when the electromagnetic interaction proceeds in the single-particle regime. This means that the energy-momentum excess during scattering is transferred to one plasma electron as in Compton scattering of X-radiation by an atom, when an atomic electron is knocked out of the atom, taking away the energy-momentum excess. Corresponding to Compton scattering in the plasma is the first summand on the right-hand side of (A.19), describing the normalized electron dynamic form factor.

For transition scattering, the situation is quite the opposite: the inequation $|\Delta \mathbf{k}| < r_D^{-1}$ is satisfied, implying that scattering of an electromagnetic field occurs by a Debye sphere surrounding an ion in the plasma as by an unit. In this case the energy-momentum excess is transferred to the plasma ion.

Using (2.68) and the explicit expression for the plasma DFF (see Appendix III), then averaging over the polarization of incident radiation and integrating with respect to the scattered radiation frequency, we obtain the following angular distribution (in terms of one ion) for the probability of transition scattering of an ultrashort pulse:

$$\frac{dW_i}{N_i d\Omega'} \simeq \frac{c E_0^2 r_e^2}{8 \pi^2} Z_i^2 (1 + \cos^2 \theta) \int_0^\infty \frac{|g(\omega')|^2 d\omega'}{\hbar \omega' [1 + (2 r_D (\omega'/c) \sin(\theta/2))^2]^2}, \quad (2.79)$$

where $N_i = n_i \delta V$ is the number of ions in the scattering volume δV .

The analogous expression for the Compton scattering channel in terms of one-electron looks like

$$\frac{dW_e}{N_e d\Omega'} \simeq \frac{c E_0^2 r_e^2}{8 \pi^2} (1 + \cos^2 \theta) \int_0^\infty \frac{(2 r_D (\omega'/c) \sin(\theta/2))^4 |g(\omega')|^2 d\omega'}{\hbar \omega' [1 + (2 r_D (\omega'/c) \sin(\theta/2))^2]^2}, \quad (2.80)$$

where $N_e = n_e \delta V$. In deriving (2.79)–(2.80), we made the replacement

$$\frac{\exp\left(-\frac{\Delta \omega^2}{2 \Delta \mathbf{k}^2 v_T^2}\right)}{\sqrt{2\pi} v_T |\Delta \mathbf{k}|} \rightarrow \delta(\Delta \omega), \quad (2.81)$$

which is justified if $|\Delta \omega| \gg v_T |\Delta \mathbf{k}|$, as supposed here. The relation (2.81) amounts to equating the scattered frequency ω' with the frequency ω in the Fourier

expansion of the ultrashort pulse ($\omega' \cong \omega$), that is, it neglects inelastic processes during scattering.

For the total probability of scattering of an ultrashort pulse in a plasma by the transition channel, integrating the right-hand side of (2.79) with respect to the solid angle of scattering, we find

$$\frac{W_i}{N_i} \simeq \frac{c E_0^2 r_e^2}{4 \pi} Z_i^2 \int_0^\infty F_i \left[2(r_D(\omega'/c))^2 \right] \frac{|g(\omega')|^2 d\omega'}{\hbar \omega'}, \quad (2.82)$$

$$F_i(x) = 2 \frac{1+x}{x^3} \left\{ \frac{2x(1+x)}{2x+1} - \ln(2x+1) \right\}.$$

The analogous expression for Compton scattering is

$$\frac{W_e}{N_e} \simeq \frac{c E_0^2 r_e^2}{4 \pi} \int_0^\infty F_e \left[2(r_D(\omega'/c))^2 \right] \frac{|g(\omega')|^2 d\omega'}{\hbar \omega'}, \quad (2.83)$$

$$F_e(x) = 2 \frac{2x(4x^3 + 11x^2 + 15x + 6) - (4x^3 + 8x^2 + 7x + 2) \ln(2x+1)}{3x^3(2x+1)}.$$

Let us use these expressions to describe scattering of an ultrashort Gaussian pulse (2.75) in a plasma. The value of $|g(\omega')|^2$ to be included in the above expressions is given in this case by (2.76)–(2.78). In the long pulse limit $n_c \gg 1$, (2.76)–(2.78) imply

$$|g(\omega)|^2 \rightarrow \pi^2 \frac{n_c}{\omega_c} \delta(\omega - \omega_c). \quad (2.84)$$

In this case (2.82) and (2.83) simplify, and for the total probability of scattering by both channels normalized by the number of electrons $N_e = Z_i N_i$, we have

$$\frac{W(n_c \gg 1)}{N_e} \simeq \frac{\pi c E_0^2 n_c}{4 \hbar \omega_c^2} r_e^2 F \left[2(d_e(\omega_c/c))^2 \right], \quad (2.85)$$

where $F = F_e + Z_i F_i$. This analysis shows that the difference between the probability of scattering of an ultrashort pulse in a plasma as calculated using (2.82) and (2.83) and in the monochromatic limit (2.85) exists only for subcycle pulses $n_c \leq 1$. This difference increases with growing carrier frequency of the pulse and has a non-monotonic dependence on the Debye radius.

The results of calculations of the spectral and angular probability of scattering of an electromagnetic pulse in plasma are illustrated in Figs. 2.13, 2.14, 2.15, and 2.16. Plotted on the ordinate in these figures is the scattering probability for the whole time of action of a pulse normalized to the intensity $I_0 = c E_0^2 / 8 \pi$. We assume everywhere that $\omega_c > \omega_{pe}$, where ω_{pe} is the electron plasma frequency. The dependence of the spectral curve of scattering of ultrashort pulses on the CE

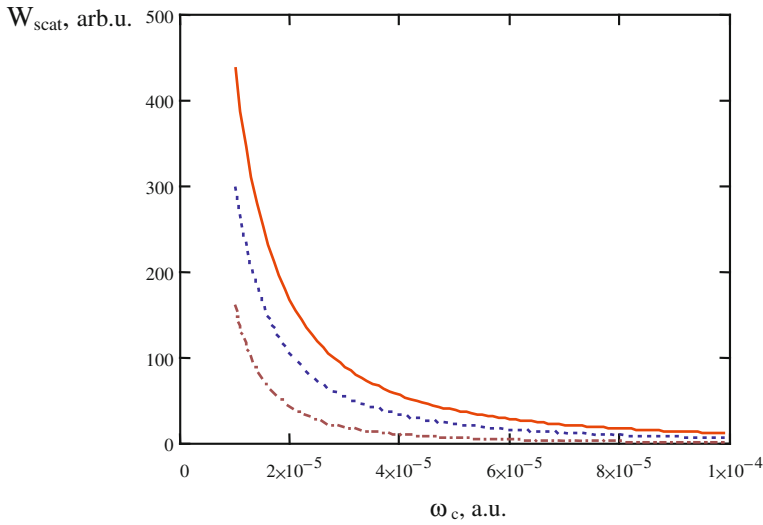


Fig. 2.13 Dependence of the normalized probability of scattering of a quarter-cycle pulse in a plasma on the carrier frequency for three values of the CE phase, $r_D = 10^5$ a.u. *solid curve* $\varphi = 0$, *dotted curve* $\varphi = \pi/4$, *dash-and-dot curve* $\varphi = \pi/2$

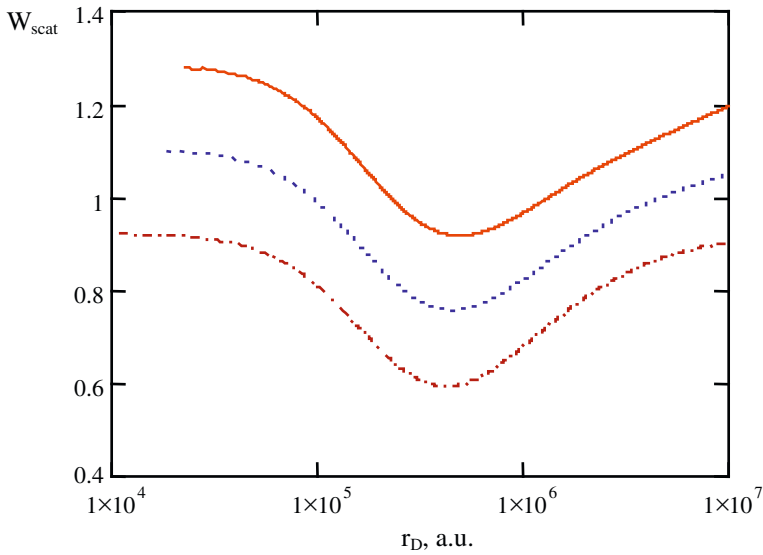


Fig. 2.14 Normalized probability of scattering of a half-cycle pulse as a function of the Debye radius for different values of the CE phase, $\omega_c = 3 \cdot 10^{-4}$ a.u. *solid curve* $\varphi = 0$, *dotted curve* $\varphi = \pi/4$, *dash-and-dot curve* $\varphi = \pi/2$

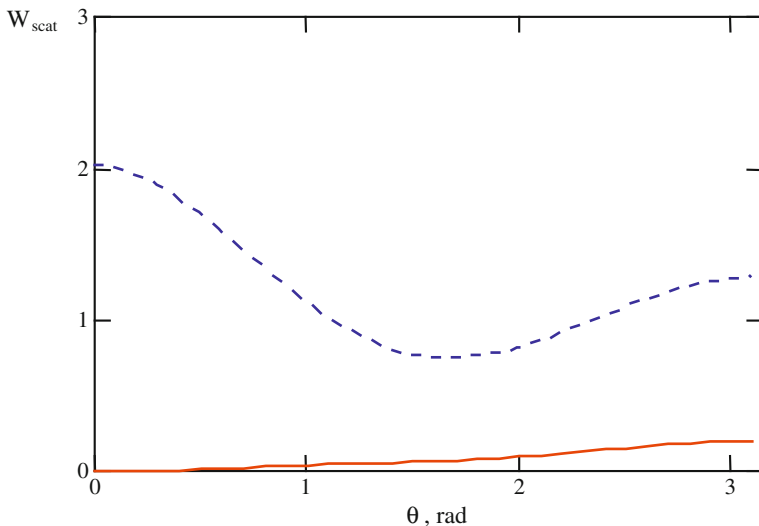


Fig. 2.15 Angular dependence of the normalized probability of scattering of a quarter-cycle pulse ($\omega_c = 10^{-4}$ a.u., $r_D = 10^6$ a.u.) by different channels: *solid curve* Compton scattering, *dotted curve* transition scattering for $Z_i = 1$

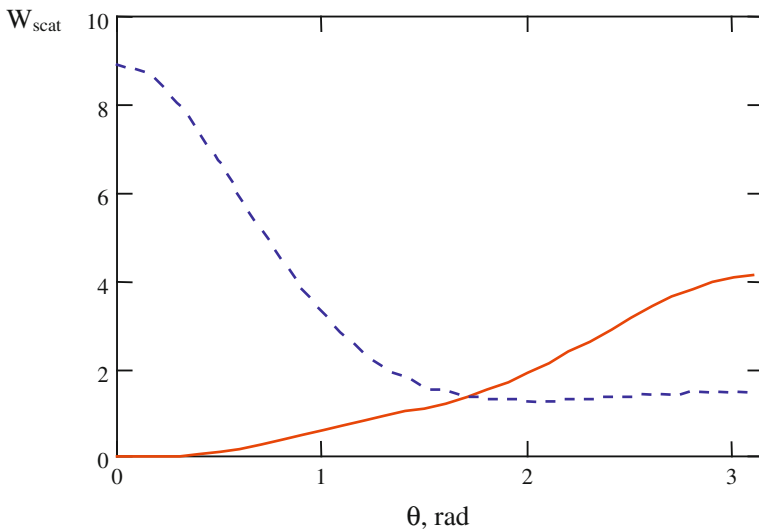


Fig. 2.16 The same as in Fig. 2.15 but for a five-cycle pulse

phase is manifest only for subcycle pulses $n_c < 1$. In this case, as can be seen from Fig. 2.13, growth of the CE phase in an interval from 0 to $\pi/2$ results in decreasing scattering probability.

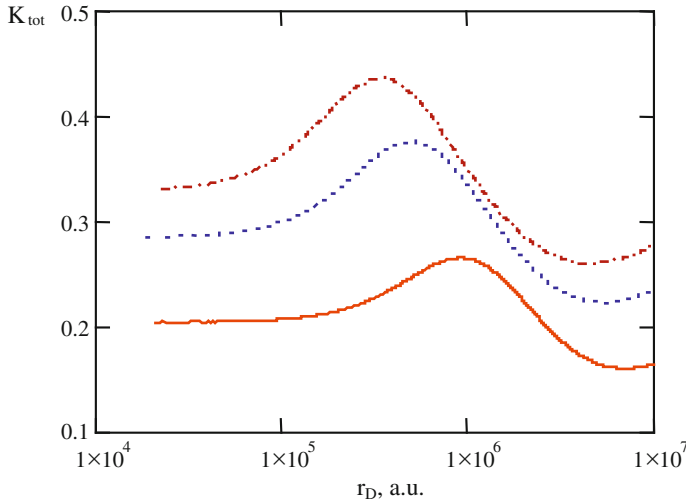


Fig. 2.17 Dependence of the phase modulation factor on the Debye radius for different values of the carrier frequency of a half-cycle pulse: *solid curve* $\omega_c = 10^{-4}$ a.u., *dotted curve* $\omega_c = 2 \cdot 10^{-4}$ a.u., *dash-and-dot curve* $\omega_c = 3 \cdot 10^{-4}$ a.u.

The dependence of the scattering probability for the whole time of action of a half-cycle pulse on the value of the Debye radius is presented in Fig. 2.14 for the carrier frequency $\omega_c = 3 \cdot 10^{-4}$ a.u. and three values of the CE phase. It can be seen that corresponding curves have a minimum near the value $r_D \approx 5 \cdot 10^5$ a.u. The calculation shows that, with decreasing carrier frequency, the minimum in the dependence of the scattering probability on the Debye radius is shifted to the region of greater values. With growing pulse duration, this minimum is weakly shifted to the region of smaller Debye radii.

Figures 2.15 and 2.16 show the values for the two process channels as a function of the angle of photon scattering for quarter-cycle (Fig. 2.15) and five-cycle (Fig. 2.16) pulses.

We see that with increasing angle the probability of Compton scattering grows, and the probability of transition scattering has a non-monotonic dependence.

From Figs. 2.15 and 2.16, it follows that, with decreasing pulse width, the relative contribution of Compton scattering of an ultrashort electromagnetic pulse in a plasma decreases in comparison with the contribution of transition scattering.

To characterize the dependence of the probability of ultrashort pulse scattering on the CE phase, it is convenient to introduce the phase modulation factor for the scattering probability by the formula

$$K_{tot} = 2 \frac{W(\varphi = 0) - W(\varphi = \pi/2)}{W(\varphi = 0) + W(\varphi = \pi/2)}. \quad (2.86)$$

The results for calculations of this value for scattering of a half-cycle pulse in a plasma are shown in Figs. 2.17 and 2.18. Figure 2.17 graphs the dependence of the

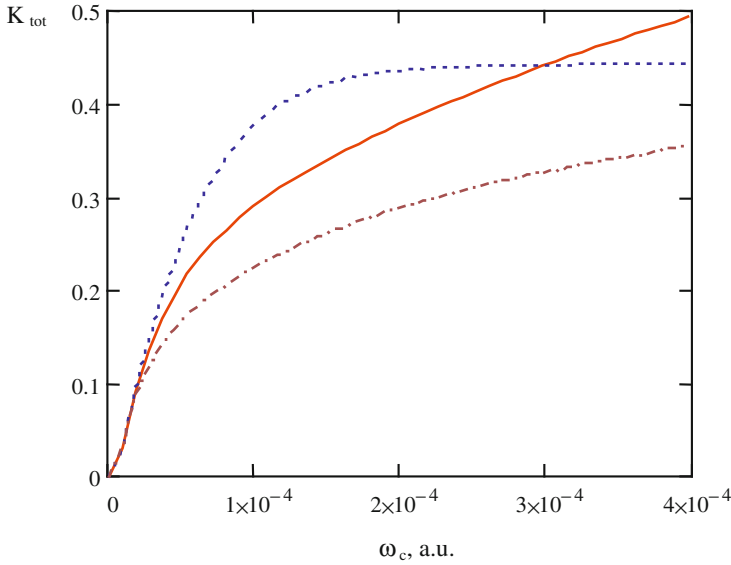


Fig. 2.18 Dependence of the phase modulation factor for scattering of a half-cycle pulse on the carrier frequency for different values of the Debye radius: *solid curve* $r_D = 10^5$ a.u., *dotted curve* $r_D = 10^6$ a.u., *dash-and-dot curve* $r_D = 10^7$ a.u.

phase modulation factor (2.86) on the value of the Debye radius for different values of the carrier frequency of a pulse. It is found that this dependence has non-monotonic behavior, the maximum being shifted to the region of smaller radii with increasing carrier frequency. Furthermore, from Fig. 2.17 we see that the phase dependence of the scattering probability grows with increasing carrier frequency of the ultrashort pulse. The same fact follows from the plots of Fig. 2.18, in which the phase modulation factor is presented as a function of the carrier frequency for different values of the Debye radius.

The above analysis thus shows that the dependence of the probability of scattering of an ultrashort electromagnetic pulse on the Debye radius in plasma has non-monotonic behavior. The minimum of the pulse scattering probability is shifted to the region of smaller values of the Debye radius with decreasing carrier frequency and increasing pulse duration.

From the analysis of angular dependences of two channels of scattering of an ultrashort pulse in plasma, it follows that, with decreasing pulse duration, the contribution of Compton scattering to the total probability of the process decreases, and in this case the probability of transition scattering for wide angles increases.

It is found that the phase dependence of scattering is manifest only for subcycle pulses $n_c < 1$, with growth of the CE phase in an interval from 0 to $\pi/2$ resulting in decreasing process probability, and increased carrier frequency resulting in increasing phase modulation factor (2.86).

A.1 2.5 Appendix III Dynamic Form Factor of Plasma Particles

The dynamic form factor (DFF) defines the probability of electromagnetic interactions with participation of plasma particles, during which the subsystem of plasma electrons or ions absorbs the energy–momentum excess. Such processes are exemplified by radiation scattering in plasma, bremsstrahlung and polarization bremsstrahlung on plasma particles including the stimulated bremsstrahlung effect, and a number of other phenomena.

The determination of the DFF of a specified plasma component has the form

$$S(\omega, \mathbf{k}) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt e^{i\omega t} \langle \hat{n}(\mathbf{k}, t) \hat{n}(-\mathbf{k}) \rangle, \quad (\text{A.1})$$

where $\hat{n}(\mathbf{k})$, $\hat{n}(\mathbf{k}, t)$ are spatial Fourier transforms of the operators representing the concentration of plasma particles of a specified type in the Schrödinger and Heisenberg pictures, and the angle brackets include both quantum–mechanical and statistical averages.

It will be recalled that the Heisenberg representation of quantum–mechanical operators takes into account their time dependence, in contrast to the Schrödinger representation, in which the whole time dependence is transferred to the wave function of the system. The relationship between these representations for an arbitrary operator \hat{Q} is

$$\hat{Q}(t) = \exp(i\hat{H}t/\hbar) \hat{Q} \exp(-i\hat{H}t/\hbar),$$

where \hat{H} is the Hamiltonian of the quantum–mechanical system. In this appendix, however, the quantum–mechanical formalism will not be used, and the quantum description is given only for completeness. Equation (A.1) can be obtained from the formula

$$S(\omega, \mathbf{k}) = \sum_{f,i} w(i) \delta(\omega + \omega_{fi}) |n_{fi}(\mathbf{k})|^2, \quad (\text{A.2})$$

averaging over initial states $|i\rangle$ and summing over final states $|f\rangle$ of the plasma particles ($w(i)$ is the probability of a plasma particle being in the i th state). As usual the delta function in (A.2) reflects energy conservation.

Depending on the type of plasma particles, the DFF can be electronic, ionic, or mixed. For the mixed DFF, the product of the density operators for electrons and ions appears in the determination of (A.1).

Physically, the DFF defines the probability of plasma absorption of the four-dimensional wave vector $k = (\omega, \mathbf{k})$ in terms of the action of an external disturbance *on a specified plasma component*. When the charge distribution in the plasma is uniform, this probability is equal to zero, since then the Fourier

transform of the density of charged particles reduces to the delta function $n(\mathbf{k}) \rightarrow n \delta(\mathbf{k})$. Thus the DFF is connected with charge fluctuations in the plasma.

In fact, the dynamic form factor reflects the dynamics of plasma particles interacting with each other through long-range Coulomb forces. Interactions are then taken into account within the ensemble of one type of particles and also between electrons and ions.

For an uniform plasma, it is convenient to introduce the DFF of the unit volume (the normalized DFF) by the formula

$$\tilde{S}(\omega, \mathbf{k}) = \frac{S(\omega, \mathbf{k})}{V}, \quad (\text{A.3})$$

where V is the volume of the plasma. This equation follows from the fact that, for an uniform medium, the pair correlation function of the concentration depends only on the relative distance between spatial points:

$$Kn(\mathbf{r}, \mathbf{r}', t) \equiv \langle \hat{n}(\mathbf{r}, t) \hat{n}(\mathbf{r}', 0) \rangle = Kn(\mathbf{r} - \mathbf{r}', t).$$

To calculate the normalized DFF, it is convenient to use the fluctuation–dissipation theorem connecting the DFF of a plasma component with the function describing the plasma response to the external electromagnetic disturbance [8]. This theorem for the electron DFF is expressed by

$$\tilde{S}_e(\omega, \mathbf{k}) = \frac{\hbar}{\pi e^2} \frac{\text{Im}\{F_{ee}(\omega, \mathbf{k})\}}{[\exp(-\hbar\omega/T) - 1]} \quad (\text{A.4})$$

where $F_{ee}(\omega, \mathbf{k})$ is the linear function describing the electron component response to the fictitious external potential acting only on plasma electrons, and T is the temperature of the plasma in energy units. The imaginary part of the response function appearing in (A.4) describes energy dissipation in the plasma, whence the name for the theorem.

With reference to [8], we introduce a second linear function describing the response to the external potential $F_{ei}(\omega, \mathbf{k})$, i.e., describing the response of the electron component of the plasma to the action of the fictitious external potential acting only on plasma ions. Here for convenience we use the Coulomb gauge of the electromagnetic field, in which the divergence of the vector potential is equal to zero ($\text{div}\mathbf{A} = 0$) and the charge density is related only to the scalar potential of the electromagnetic field φ via the Poisson equation. So the external potential $\varphi_{ext}(k)$ acts on the plasma, where $k = (\omega, \mathbf{k})$ is the four-dimensional wave vector. Then the density of the electron charge induced in the plasma is expressed in terms of the above response functions as follows:

$$\langle \hat{\rho}_e(k) \rangle = [F_{ee}(k) + F_{ei}(k)] \varphi_{ext}(k). \quad (\text{A.5})$$

$\langle \hat{\rho}_j(k) \rangle = e_j \langle \hat{n}_j(k) \rangle$ is the charge density of the j th type of plasma particles. Equation (A.5) indicates that the electron charge density arises in the plasma due to direct action of the external potential on plasma electrons [the first summand in

the square brackets of (A.5)] and also as a result of the action of the external potential on plasma ions that are bound to electrons by Coulomb forces. If interaction between particles of type i and type j is weak, one can use the technique described in [8] to express F_{ij} in terms of the characteristics of noninteracting particles. For this purpose the new response function $\alpha_j(k)$ is introduced—the response function for particles of type j to the *total potential* in the plasma. It takes into account the action on charged particles of the potential $\varphi_{ind}(k)$ induced in the plasma due to redistribution of the charged particles under the action of the external potential. With the help of the function $\alpha_j(k)$, the induced charge density for the j th component can be expressed in terms of the total potential:

$$\langle \hat{\rho}_j(k) \rangle = \alpha_j(k) \varphi_{tot}(k). \quad (\text{A.6})$$

As the response function $\alpha_j(k)$ describes the action of the total potential on the plasma particles, the characteristics of noninteracting particles can be used to calculate it, since interaction between them is already taken into account in the total potential. This technique is widely used in plasma physics to describe screening and initiation of collective excitations. In the approach under consideration, corresponding to the random phase approximation [8], $\alpha_j(k)$ can be expressed in terms of the function $Q_j(k)$ characterizing the noninteracting particles according to $\alpha_j = e_j^2 Q_j$, where

$$Q_j(k) = \int \frac{n_j(\mathbf{p} + \hbar \mathbf{k}) - n_j(\mathbf{p})}{E_j(\mathbf{p} + \hbar \mathbf{k}) - E_j(\mathbf{p}) - \hbar \omega - i0} \frac{2 d\mathbf{p}}{(2\pi\hbar)^3}. \quad (\text{A.7})$$

Here $n_j(\mathbf{p})$ is the dimensionless momentum distribution function of plasma particles of type j and $E_j(\mathbf{p}) = p^2/2m_j$. Hereafter we need to know the imaginary part of the function $Q_j(k)$, which can be determined from (A.7) using the Sokhotsky formula. For the Maxwell velocity distribution of the electrons, we find

$$\text{Im}\{Q_j(k)\} = \pi (e^{-\hbar\omega/T} - 1) n_j \frac{\exp\left\{-\omega^2/2k^2 v_{Tj}^2\right\}}{\sqrt{2\pi} k v_{Tj}}. \quad (\text{A.8})$$

The functions introduced above to describe the response to the total potential are related to the longitudinal part of the dielectric permittivity by

$$\varepsilon^{(lj)}(k) = 1 - \frac{4\pi}{k^2} \alpha_j(k). \quad (\text{A.9})$$

We can now solve the original problem, i.e., we will find the function $F_{ee}(\omega, \mathbf{k})$ and express it in terms of the function describing the response to the total potential. For this purpose we introduce the fictitious external potential φ_{ext}^* acting only on electrons. Then according to the definition of $F_{ee}(\omega, \mathbf{k})$, we have

$$\langle \hat{\rho}_e^*(k) \rangle = F_{ee}(k) \varphi_{ext}^*(k). \quad (\text{A.10})$$

On the other hand, $\langle \hat{\rho}_e^* \rangle$ can be expressed in terms of α_e :

$$\langle \hat{\rho}_e^*(k) \rangle = \alpha_e(k) [\varphi_{ext}^*(k) + \varphi_{ind}^*(k)], \quad (\text{A.11})$$

where φ_{ind}^* is the potential induced under the action of φ_{ext}^* , determined in terms of the density of all plasma charges with the help of the Poisson equation:

$$\varphi_{ind}^*(k) = \frac{4\pi}{\mathbf{k}^2} [\langle \hat{\rho}_e^*(k) \rangle + \langle \hat{\rho}_i^*(k) \rangle]. \quad (\text{A.12})$$

Here

$$\langle \hat{\rho}_i^*(k) \rangle = \alpha_i(k) \varphi_{ind}^*(k), \quad (\text{A.13})$$

Since the potential φ_{ext}^* is assumed to act only on electrons. Solving the system of Eqs. (A.8–A.12), we find the following expression for F_{ee} :

$$F_{ee}(k) = \frac{\alpha_e(k) [1 - (4\pi/\mathbf{k}^2) \alpha_i(k)]}{1 - (4\pi/\mathbf{k}^2) [\alpha_e(k) + \alpha_i(k)]}. \quad (\text{A.14})$$

Substituting (A.13) into (A.4) and using (A.8) and (A.7), we obtain

$$\tilde{S}_e(k) = \left| \frac{\varepsilon^{l(i)}(k)}{\varepsilon^l(k)} \right|^2 |\delta n_e(k)|^2 + z_i^2 \left| \frac{1 - \varepsilon^{l(e)}(k)}{\varepsilon^l(k)} \right|^2 |\delta n_i(k)|^2, \quad (\text{A.15})$$

where

$$|\delta n_{e,i}(k)|^2 = \frac{n_{e,i}}{\sqrt{2\pi} v_{Te} |\mathbf{k}|} \exp\left(-\frac{\omega^2}{2\mathbf{k}^2 v_{Te}^2}\right) \quad (\text{A.16})$$

are the spatio-temporal Fourier transforms of the squared thermal fluctuations of the electron and ionic components of the plasma calculated for the four-dimensional wave vector $k = (\mathbf{k}, \omega)$. z_i is the charge number of the plasma ions and it is implied that the quasi-neutrality condition is satisfied, so that $n_e = z_i n_i$.

The expression for the normalized ionic DFF is found in exactly the same way as the electron DFF. For this purpose, one must make the index replacement $e \rightleftharpoons i$ and take into account the fact that, in the denominator of (A.4), the ion charge $e_i = z_i e$ now appears. Then we obtain:

$$\tilde{S}_i(k) = \left| \frac{\varepsilon^{l(i)}(k)}{\varepsilon^l(k)} \right|^2 |\delta n_i(k)|^2 + z_i^{-2} \left| \frac{1 - \varepsilon^{l(i)}(k)}{\varepsilon^l(k)} \right|^2 |\delta n_e(k)|^2. \quad (\text{A.17})$$

The mixed normalized DFF is given by

$$\tilde{S}_{ei}(k) = z_i^{-1} \left| \frac{1 - \varepsilon^{l(i)}(k)}{\varepsilon^l(k)} \right|^2 |\delta n_e(k)|^2 + z_i \left| \frac{1 - \varepsilon^{l(e)}(k)}{\varepsilon^l(k)} \right|^2 |\delta n_i(k)|^2, \quad (\text{A.18})$$

which follows from the fluctuation–dissipation theorem (A.4) (with the replacement $e^2 \rightarrow e e_i$) and the formula for the linear response function F_{ei} describing the initiation of an electron charge induced by the fictitious potential that acts only on ions. This formula has the form

$$F_{ei}(k) = \frac{(4\pi/k^2) \alpha_i(k) \alpha_e(k)}{1 - (4\pi/k^2) [\alpha_e(k) + \alpha_i(k)]}. \quad (\text{A.19})$$

Equation (A.18) is obtained by analogous reasoning to the deduction of (A.13).

Let us explain the physical meaning of the expression (A.14) for the electron DFF. The first summand is connected with the deficiency of electron charge around the electron density fluctuation, caused by electron–electron repulsion. The second summand in this expression describes the electron charge screening the fluctuation of the ionic plasma component. It results from electron–ion attraction. By analogy, in the expression (A.16) for the ionic DFF, the second summand describes the ionic charge screening the electron density fluctuation, while the first summand describes the deficiency of ionic charge around the ionic fluctuation. Finally, in the formula (A.16) for the mixed DFF, the first summand describes the ionic charge screening the electron density fluctuation, and the second summand describes the electron charge screening the ionic density fluctuation.

Let us consider the explicit form of the electron DFF satisfying the inequations $k v_{Te} \gg \omega \gg k v_{Ti}, \omega_{pi}$. Then the low frequency approximation is valid for the longitudinal electron dielectric permittivity of the plasma, and the high-frequency approximation is valid for the ionic component. Using the expressions for the longitudinal part of the dielectric permittivity of the plasma and the formula (A.15), we find

$$\tilde{S}_e(k) \simeq \left(\frac{k^2 r_{De}^2}{1 + k^2 r_{De}^2} \right)^2 |\delta n_e(k)|^2 + \frac{z_i^2}{(1 + k^2 r_{De}^2)^2} |\delta n_i(k)|^2. \quad (\text{A.20})$$

From this formula we see that, in the case of long-wavelength fluctuations, when $k^2 r_{De}^2 \ll 1$ ($k = 2\pi/\lambda$), the first summand describing the deficiency of electron charge around the electron density fluctuation is small. The second summand connected with electron screening of ionic density fluctuations is large. Hence it follows that, in the long-wavelength limit, the transfer of energy–momentum to the plasma proceeds through the electron charge of the Debye sphere around a plasma ion which reacts in a coherent manner to the electric field. That is, interaction is of a collective nature. In the short-wavelength case $k^2 r_{De}^2 \gg 1$, the situation is opposite: electromagnetic interaction is realized through excitation of individual plasma electrons, under which the Debye sphere “falls apart” due to the strong spatial non-uniformity of the electric field.

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Matter

Astapenko, V.

2013, VIII, 94 p. 44 illus., 40 illus. in color., Softcover

ISBN: 978-3-642-35968-2