Quantum Electronics **30** (11) 979 – 985 (2000)

NONLINEAR OPTICAL PHENOMENA

PACS numbers: 42.50.Hz; 42.65.Ky; 42.65.Re
DOI: 10.1070/QE2000v030n11ABEH001848

The influence of the spatial inhomogeneity of the field on the nonlinear-optical response of an atom

A V Andreev, A B Kozlov

Abstract. The theory of the interaction of a centrosymmetric atom with a superstrong spatially inhomogeneous laser field is developed. This theory employs the two-level approximation to describe the dynamics of spatially nonlocal interactions related to variations in the populations of the levels. We propose a model that includes spatially nonlocal interactions, such as magnetic-dipole and quadrupole interactions and interactions due to the gradient of the ponderomotive potential of the field. We consider the interaction of a homogeneous medium of centrosymmetric atoms with a superstrong laser field, which is represented as a superposition of two plane-wave ultrashort pulses propagating at some angle with respect to each other. Perturbation theory, which is valid for the fields of moderate intensity, is developed for the atomic response. The results of numerical simulations are compared with the predictions of this perturbation theory. The specific features of the nonlinear-optical response of an atom in a superstrong field are investigated. The angular distribution of the second- and third-harmonic emission is calculated in the constant-field approximation for different polarisations of the incident field.

1. Introduction

In recent years, a permanent interest has been expressed in the physics of propagation of ultrahigh-power femtosecond light pulses and the interaction of these pulses with both single atoms and dense media. This interest is associated with nonlinear-optical processes that cannot be described within the framework of conventional approximations of nonlinear optics [1 – 5]. The current level of optical technologies allows the generation of laser pulses with a duration less than 5 fs [6], which corresponds to a few optical cycles.

Nonlinear-optical effects in isotropic media forbidden due to the symmetry properties of a medium have been extensively studied in the last few years [7]. This class of effects includes, for example, second-harmonic generation (SHG). As is well known, SHG is forbidden in the electric-dipole approximation in media with a central symmetry [8]. However, if a spatially inhomogeneous field, i.e., a tightly focused laser pulse [9] or two plane waves propagating at some angle with respect to each other [10], interacts with a medium, then the prohibition on SHG can be removed.

Three main mechanisms may give rise to SHG in an isotropic medium in the presence of a spatially inhomogeneous field. First, a strong field may ionise a part of atoms, resulting in a spatially nonuniform distribution of free electrons. This distribution is determined by the spatial structure of the incident field. Second-harmonic generation in a nonuniform plasma is now well understood [11, 12].

The second mechanism is referred to as dc-field-induced SHG [13 – 18]. In this regime, similar to the case considered above, a strong field ionises some part of atoms in the medium. The initial spatial distributions of electrons and ions coincide with each other. However, charges become separated in space with time. This charge separation gives rise to a macroscopic dc electric field, which induces SHG.

In this paper, we investigate the third mechanism of SHG in a spatially inhomogeneous field. The second harmonic can be generated due to spatially nonlocal atom–field interactions. The phase-matching condition, providing efficient energy conversion from the fundamental wave to the second harmonic, can be satisfied in this case due to the spatially inhomogeneous structure of the external field. Second-harmonic generation becomes possible in such a situation due to the fact that a superstrong spatially inhomogeneous field changes the symmetry of wave functions of atomic electrons.

The third mechanism dominates over the first two mechanisms if the ionisation probability of atoms in the medium is low and the pulse duration is much less than the build-up time of the induced dc field. Several experimental and theoretical studies have confirmed the possibility of generating the second harmonic due to spatially nonlocal interactions.

Second-harmonic generation accompanying the noncollinear interaction of two waves was considered in Ref. [19]. The authors of [19] found that the highest efficiency of SHG is achieved in the direction of the bisectrix of the angle between the wave vectors of the pump waves. The second harmonic is s-polarised in this case. Below, we will show that these predictions are valid only within the framework of perturbation theory, when population variations are ignored.

Generally, the amplitude of the second harmonic depends also on the polarisations of the waves interacting with the medium. The theory of interaction of an atom with a superstrong spatially inhomogeneous field developed in [20] was employed to explain the results of experiments on SHG in spatially periodic media [10].

This paper considers the interaction of a homogeneous medium of centrosymmetric atoms with a superstrong spa-

A V Andreev, A B Kozlov International Teaching and Research Laser Centre, M V Lomonosov Moscow State University, Vorob'evy gory, 119899 Moscow, Russia

Received 17 March 2000
Kvantovaya Elektronika **30** (11) 979 – 985 (2000)
Translated by A M Zheltikov

tially inhomogeneous field, which can be represented as a superposition of two plane-wave pulses propagating at an angle to each other. Our analysis involves equations for the field that are free of the assumptions of the slowly varying envelope approximation and equations for the medium written in the two-level approximation.

In the case of ultrashort light pulses with a moderate intensity, when ionisation is negligible, the ground and first excited levels coupled with each other by a dipole-allowed transition have the largest populations. The influence of other levels is included in the model within the framework of perturbation theory.

Our model will include spatially nonlocal interactions (magnetic-dipole and quadrupole interactions, as well as interactions due to the gradient of the ponderomotive potential of the field) with an accuracy up to the first spatial derivative of the vector potential of the field. The set of equations for the atomic response derived within the framework of this model allows both resonant and nonresonant interactions of an atom with a field pulse to be analysed. Since we abandon the approximation of slowly varying amplitudes and phases, the light pulse may also have an arbitrary duration.

The main advantage of the approach proposed in this paper over the conventional perturbation theory is that this approach allows us to investigate the dynamics of the atomic response related to the evolution of level populations in the process of interaction. The contributions of different spatially inhomogeneous interactions change in time, which implies that we deal with a nonstationary response of an atom.

2. Equations governing the dynamics of the atomic response

The response of an atom to an external electromagnetic field can be conveniently characterised with a set of variables defined as quantum-mechanical means of the Hamiltonian $\hat{H}_0$ of a free atom and operators of the atomic dipole moment $\hat{\boldsymbol{d}} = e\hat{\boldsymbol{r}}$, the canonical electron momentum $\hat{\boldsymbol{p}}$, and the electron velocity $\hat{\boldsymbol{v}}$. Operators of current density $\hat{\boldsymbol{j}} = e\hat{\boldsymbol{p}}/m$ and $\hat{\boldsymbol{J}} = e\hat{\boldsymbol{v}}$ are usually introduced instead of the operators of the electron momentum and velocity.

Thus, we deal with the following set of variables:

$$\boldsymbol{d} = \int \Psi^* \hat{\boldsymbol{d}} \Psi \mathrm{d}V, \; \boldsymbol{j} = \int \Psi^* \hat{\boldsymbol{j}} \Psi \mathrm{d}V, \; \boldsymbol{J} = \int \Psi^* \hat{\boldsymbol{J}} \Psi \mathrm{d}V,$$

$$E = \int \Psi^* \hat{H}_0 \Psi \mathrm{d}V, \qquad (1)$$

where E is the energy of the electron subsystem of an atom. The difference of current densities $\boldsymbol{j}$ and $\boldsymbol{J}$ in the dipole approximation is proportional to the vector potential $\boldsymbol{A}$ of the field. Spatially nonlocal interactions of an atom with an external field give rise to differences in the dynamics of current densities $\boldsymbol{j}$ and $\boldsymbol{J}$.

The change in the vector potential of the field within characteristic sizes of orbits of atomic electrons will be included in our approach with an accuracy up to the first derivative. In other words, spatially nonlocal interactions, such as magnetic-dipole and quadrupole interactions, as well as interactions due to the gradient of the ponderomotive potential of the field, will be included in our analysis along with dipole interactions. We should note that the variables $\boldsymbol{d}$ and $\boldsymbol{j}$ in a two-level atom are proportional to two quadrature components of polarisation.

Consider the atomic response of a medium within the framework of the model of a two-level atom. In this case, we have $E = R\hbar\omega_0/2$, where R is the population difference for atomic levels and ω_0 is the frequency of the atomic transition. Because of selection rules for matrix elements of transitions in a centrosymmetric atomic potential, the model of a two-level atom does not allow us to include all the spatially nonlocal interactions of an atom with an external field. If two levels are coupled by an electric-dipole transition, then magnetic-dipole and quadrupole transitions between these levels are forbidden. Thus, the model of a two-level atom with electric-dipole transitions includes only dipole interactions and interactions due to the gradient of the ponderomotive potential of the field.

To include all the spatially nonlocal interactions in a correct way, we should generalise the model of a two-level atom. Consider two atomic energy states coupled by an electric-dipole-allowed transition. Suppose that the atom can be found in one of these states with a probability close to unity, i.e., the sum of populations of these two levels is nearly unity. The population of all the other atomic states will be assumed to be small, but nonzero.

Let us examine the changes in the population of these states due to magnetic-dipole and quadrupole interactions of an atom with the external field. The probability of transitions induced by these interactions is low as compared to the probability of the considered electric-dipole transition. Therefore, we can neglect changes in the atomic energy due to the above-specified spatially nonlocal interactions, thus assuming that magnetic-dipole and quadrupole interactions may change the polarisation of an atom, but never change the atomic energy. The Stark shift of atomic levels plays an important role in the interaction of a superstrong field with an atom. This effect will be included within the framework of perturbation theory.

Consider the electromagnetic field propagating in a homogeneous medium with a concentration of atoms equal to N/V. With the assumptions specified above, the set of equations for the vector potential $\boldsymbol{A}$ of the field and atomic variables (1) can be written as

$$\mathrm{rot}\,\mathrm{rot}\,\boldsymbol{A} + \frac{1}{c^2}\frac{\partial^2 \boldsymbol{A}}{\partial t^2} = \frac{4\pi}{c}\frac{N}{V}\boldsymbol{J},$$

$$\frac{\partial d_\alpha}{\partial t} = \left(1 - \frac{2|d|^2}{\hbar^2 c^2}\boldsymbol{A}^2\right) j_\alpha + \frac{2|d|^2\omega_0}{\hbar c} R A_\alpha - \frac{e}{mc} d_\beta \nabla_\beta A_\alpha,$$

$$\frac{\partial j_\alpha}{\partial t} = -\omega_0^2\left(1 - \frac{2|d|^2}{\hbar^2 c^2}\boldsymbol{A}^2\right) d_\alpha + \frac{e}{mc} j_\beta \nabla_\alpha A_\beta + \frac{e\omega_0 |d|^2}{mc^2\hbar} R \nabla_\alpha \boldsymbol{A}^2, \qquad (2)$$

$$\frac{\partial R}{\partial t} = -\frac{2\omega_0}{\hbar c} d_\alpha A_\alpha - \frac{e}{\hbar\omega_0 m c^2} j_\alpha \nabla_\alpha \boldsymbol{A}^2,$$

$$\boldsymbol{J} = \boldsymbol{j} - \frac{e^2}{mc}\boldsymbol{A} - \frac{e}{mc}(\boldsymbol{d}\nabla)\boldsymbol{A},$$

where $|d|$ is the matrix element of the dipole moment of the transition; e and m are the charge and the mass of an electron; c is the speed of light; and α and β are the coordinate indices, which take the values x, y, and z. Since we abandon the approximation of slowly varying amplitudes and phases, the set of equations (2) allows us to investigate both resonant and nonresonant interactions of a two-level atom with an ultrashort pulse of the electromagnetic field with an arbitrary duration.

3. Perturbation theory

Consider the specific features of the atomic nonlinear-optical response caused by the spatial inhomogeneity of the external field in terms of stationary perturbation theory, i.e., assuming that the population of atomic levels remains unchanged. We will assume that none of the harmonics of the external field is resonant with the frequency of the atomic transition.

The ratio of the Rabi frequency $\Omega_{\rm R} = 2|d|E_0/\hbar$ (where E_0 is the amplitude of the electric field strength) to the frequency ω_0 of the atomic transition will be taken as a small parameter of perturbation theory. Such an approach is quite reasonable if the amplitude of the external field is much less than the amplitude of the intraatomic field.

Suppose that an atom interacts with a field that can be represented as a superposition of two plane waves propagating at an angle of 2θ to each other. We choose a system of coordinates where the wave vectors can be written as $\boldsymbol{k}_1 = \{0, k\sin\theta, k\cos\theta\}$ and $\boldsymbol{k}_2 = \{0, -k\sin\theta, k\cos\theta\}$ (see Fig. 1). Generally, both waves are elliptically polarised:

$$\boldsymbol{A}_1 = \frac{A_0}{2}(\boldsymbol{e}_x a\cos\Phi_1 + \boldsymbol{e}_{y1} b\sin\Phi_1),$$
$$\boldsymbol{A}_2 = \frac{A_0}{2}(\boldsymbol{e}_x c\cos\Phi_2 + \boldsymbol{e}_{y2} d\sin\Phi_2), \qquad (3)$$

where A_0 is the amplitude of the vector potential; a, b, c, and d are the dimensionless constants describing polarisation ellipses; and $\boldsymbol{e}_{y1}$ and $\boldsymbol{e}_{y2}$ are the unit vectors related to the unit vectors of the initial system of coordinates by the expressions $\boldsymbol{e}_{y1} = \boldsymbol{e}_y\cos\theta - \boldsymbol{e}_z\sin\theta$ and $\boldsymbol{e}_{y2} = \boldsymbol{e}_y\cos\theta + \boldsymbol{e}_z\sin\theta$. The phases of the first and second waves are given $\Phi_1 = \omega t - \boldsymbol{k}_1\boldsymbol{r} + \delta$ and $\Phi_2 = \omega t - \boldsymbol{k}_2\boldsymbol{r} - \delta$, respectively, where δ is an arbitrary phase shift and ω is the frequency of the external field.

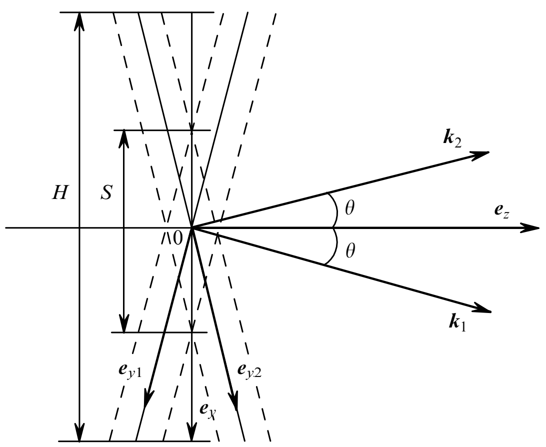

Figure 1. Geometry of the external field.

In what follows, we will employ a conventional terminology: the s component of the vector will be understood as the component perpendicular to the plane of the wave vectors, while the p component will be defined as the component lying in the plane of the wave vectors. Thus, the constants a and c determine the s component of the external field, while the constants b and d control the p component of the external field.

A spatially inhomogeneous filed interacting with an atom changes its symmetry properties. The violation degree of the central symmetry of an unperturbed atom can be characterised by a permanent dipole moment induced by the external field. Integrating Eqs. (2) and using the assumptions specified above, we can readily derive an expression for the constant dipole moment which have the only the transverse nonzero p component:

$$d_y(0) = \frac{e}{2mc^2}\frac{k\omega^2|d|^2R_0A_0^2}{\hbar\omega_0(\omega_0^2-\omega^2)}\sin\theta(ac + bd\cos 2\theta)\sin 2\xi, \quad (4)$$

where R_0 is the initial population inversion and $\xi = k\sin\theta y - \delta$. The dipole moment is modulated in the transverse coordinate, and its sign depends on the ratio of the frequency of the external field to the frequency of the atomic transition. Note that the interference of two waves is necessary to induce a permanent dipole moment of an atom.

Let us examine the response field of a medium consisting of centrosymmetric atoms at the frequency of the second harmonic. Using Eqs. (2), we can easily derive expressions for the current density at the frequency of the second harmonic. Then, substituting these expressions into the wave equation and integrating this equation in the constant-field approximation, we can find the response field of a homogeneous medium with a length L at the frequency of the second harmonic. The p component of the second harmonic is equal to zero regardless of the polarisation of the external field. The expression for the s component of the second harmonic is written as

$$E_x(2\omega) = -\frac{4\pi}{c^2k_2}\frac{N}{V}\frac{e}{mc^2}\frac{k\omega^4\omega_0|d|^2R_0A_0^2\sin 2\theta}{\hbar(\omega_0^2-\omega^2)(\omega_0^2-4\omega^2)}$$
$$\times(ad-bc)L\frac{\sin\Delta L}{\Delta L}\sin(2\omega t - k_2 z), \qquad (5)$$

where k_2 is the modulus of the wave vector of the second harmonic and $\Delta = k_2 - 2k\cos\theta$ is the phase mismatch. One can see from Eqn (5) that the s-polarised second harmonic propagates along the bisectrix of the angle between the wave vectors of two waves interacting with the medium.

The amplitude of the second harmonic depends on the polarisation of the external field. Specifically, if two s-polarised or two p-polarised waves interact with a medium, then the response field at the frequency of the second harmonic is equal to zero. Conversely, if one s-polarised and one p-polarised waves interact with a medium, then the second-harmonic response field differs from zero. When two circularly polarised waves interact with a medium, the rotation direction of the electric field vector plays an important role. If the incident waves are characterised by the same rotation direction of the electric field vector, then the second-harmonic response field vanishes. Otherwise, the response field at the frequency of the second harmonic differs from zero.

4. A two-level atom in a superstrong field

Consider the specific features of the nonlinear-optical response of a two-level atom interacting with a superstrong external field. The term 'superstrong' indicates that the Rabi frequency of the field is of the order of the frequency of the atomic transition. Suppose that the external field can be represented as a superposition of two s-polarised plane waves propagating at an angle to each other. We assume that all the harmonics of external field are not resonant with the frequency of the atomic transition.

Numerical simulation of the set of equations (2) will be performed for the following parameters: $R_0 = -1$, i.e., the atom is not excited before the interaction with a field pulse; the angle is $\theta = \pi/6$; the pulse duration at the e^{-1} level corresponds to 15 field cycles; and the difference of the phases of interacting waves is $2\delta = \pi/2$, which corresponds to the maximum transverse gradient of the ponderomotive potential of the field. Calculations were carried out for two different frequencies of the external field. In the first case, the field frequency was lower than the frequency of the atomic transition ($\omega/\omega_0 = 0.75$). In the second case, the field frequency exceeded the frequency of the atomic transition ($\omega/\omega_0 = 1.25$).

Computer simulations of the atomic response suggest that all the odd harmonics of the atomic response are s polarised, while all the even harmonics are p-polarised. As the amplitude of the external field increases, the amplitude of the current density at the frequency of the external field deviates from the linear dependence, which is characteristic of the weak-field regime (Fig. 2a). Perturbation theory yields the following expression for the refractive index of the medium:

$$n^2(\omega) = 1 - \frac{4\pi}{c}\frac{N}{V}\frac{c^2}{\omega^2}\left[\frac{e^2}{mc} + \frac{2|d|^2\omega_0^3 R_0}{\hbar c(\omega_0^2-\omega^2)} - \frac{|d|^4\omega_0^3\omega^2(3\omega_0^2+\omega^2)R_0}{\hbar c^3(\omega_0^2-\omega^2)^3}A_0^2\cos^2\xi\right]. \quad (6)$$

This expression provides a reasonable explanation of the dependences presented in Fig. 2a.

Let us apply Eqn (6) to consider the self-focusing of a linearly polarised Gaussian beam in a homogeneous medium of two-level atoms. The condition of self-focusing can be written as

$$\frac{R_0\omega_0^2}{(\omega_0^2-\omega^2)}\frac{P}{P_0} \geqslant 1, \quad (7)$$

where P is the power of laser radiation;

$$P_0 = \frac{\hbar^3 c^3(\omega_0^2-\omega^2)^2 n_0}{16\pi(N/V)|d|^4\omega_0(3\omega_0^2+\omega^2)} \quad (8)$$

is the critical power of laser radiation, and n_0 is the unperturbed refractive index of the medium in the absence of the external field. When the dimensionless factor $R_0\omega_0^2(\omega_0^2-\omega^2)^{-1}$ in Eqn (7) is positive, we deal with self-focusing. When this factor is negative, self-defocusing occurs. Thus, a laser beam undergoes self-focusing in a nonexcited medium when the field frequency exceeds the frequency of the relevant atomic transition. For a medium consisting of two-level atoms with a concentration equal to $N/V = 10^{19}$ cm^{-3}, transition dipole moment $|d| = 1$ D, and the ratio of the field frequency to the frequency of the atomic transition $\omega/\omega_0 = 1.25$, the critical power P_0 required for the self-focusing of the beam is ~ 30 kW.

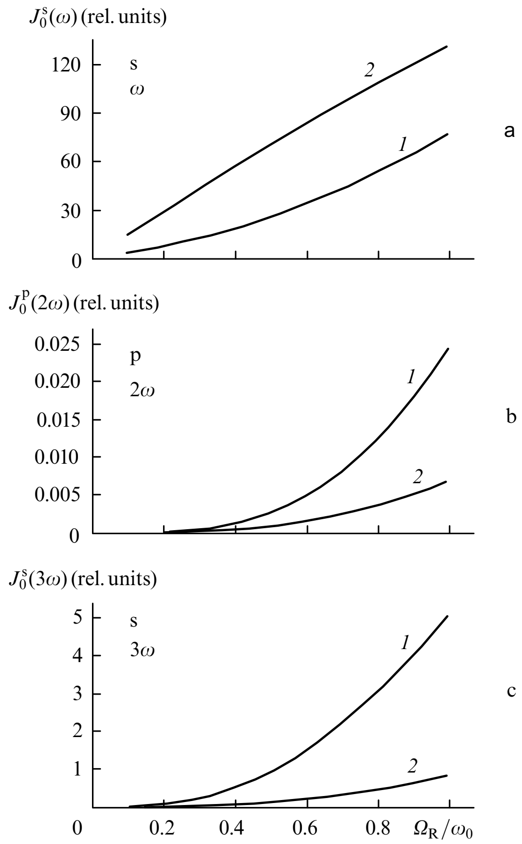

Figure 2. Amplitudes of the s component (a) of the current density at the frequency of the external field $J_0^s(\omega)$, the p component (b) of the current density at the frequency of the second harmonic $J_0^p(2\omega)$, and the s component (c) of the current density at the frequency of the third harmonic $J_0^s(3\omega)$ (c) as functions of the Rabi frequency normalised to the frequency of the atomic transition with (*1*) $\omega/\omega_0 = 0.75$ and (*2*) 1.25.

Fig. 2b displays the amplitude of the p component of the current density at the frequency of the second harmonic as a function of the Rabi frequency normalised to the frequency of the atomic transition. The second order of perturbation theory does not contribute to the atomic response at the frequency of the second harmonic. Conse-quently, the response at the frequency of the second harmonic comes from the fourth and higher orders of perturbation theory. This implies that the current density should be described by a quartic function of the amplitude of the external field in the weak-field regime.

As the amplitude of the external field increases, this dependence saturates, i.e., the exponent of the power function becomes less than four. In a strong field, the cubic dependence of the amplitude of the current density at the frequency of the third harmonic on the amplitude of the external field, which is characteristic of the weak-field regime, saturates (Fig. 2c). In other words, the exponent of the power function in this case becomes less than three.

5. The angular spectrum of the response field of a medium

Consider the interaction of a homogeneous medium of two-level atoms with a superposition field of two plane-wave pulses propagating at an angle to each other. The dashed lines in Fig. 1 show the areas whose sizes are determined by pulse durations. The transverse sizes of the medium of two-level atoms and the area where the wave packets overlap are equal to H and S, respectively.

The atomic response of a homogeneous medium of length L interacting with the field of two plane waves propagating at an angle to each other is described by a periodic function of the transverse coordinate y. Therefore, we can represent the response field in the form of a Fourier series:

$$\boldsymbol{E}^{\mathrm{s,p}} = \sum_{m=1}^{+\infty} \sum_{p=-\infty}^{+\infty} \left(\frac{1}{4q_{mp}} \frac{\sin \varDelta_{mp} L}{\varDelta_{mp}} \right) \vec{\mathscr{E}}^{\,\mathrm{s,p}}_{mp}, \tag{9}$$

where $q_{mp} = (k_m^2 - p^2 k^2 \sin^2\theta)^{1/2}$ is the z-component of the wave vector, k_m is the modulus of the wave vector of the mth harmonic, and $\varDelta_{mp} = q_{mp} - mk\cos\theta$ is the phase mismatch of the wave vectors. The index m numerates harmonics of the carrier frequency, while the index p corresponds to different propagation directions of the response field of the medium. The set of these propagation directions is discrete because of the periodic structure of the incident field.

The phase-matching condition for the mth harmonic propagating along the direction characterised by parameter p has the form

$$\left(\frac{n_m}{n}\right)^2 = 1 + \left[\left(\frac{p}{m}\right)^2 - 1\right]\sin^2\theta, \tag{10}$$

where n_m and n are the refractive indices of the medium for the mth harmonic and the incident field, respectively.

Let us introduce the angle φ_{mp} between the propagation direction of the mth harmonic, which corresponds to the parameter p, and the z-axis. Generally, the angle φ_{mp} depends on the ratio of the refractive indices of the medium at the relevant frequencies. However, if the phase-matching condition is satisfied for some propagation direction of the response field corresponding to the parameter p, then the angle φ_{mp} is determined by the expression

$$\sin\varphi_{mp} = \frac{p}{m}\sin\theta \left\{ 1 + \left[\left(\frac{p}{m}\right)^2 - 1\right]\sin^2\theta \right\}^{-1/2}. \tag{11}$$

Our numerical simulations were aimed at calculating the angular spectrum of the response field $\vec{\mathscr{E}}^{\,\mathrm{s,p}}_{mp}$ for different polarisations of the incident field. The following parameters were employed in our numerical simulations: $\theta = \pi/6$, $\omega/\omega_0 = 0.75$, $R_0 = -1$ (the medium is not excited before the interaction with the field); $\Omega_{\mathrm{R}}/\omega_0 = 0.2$; the pulse duration at the e^{-1} level corresponds to 15 field cycles; $S = 30\lambda$, where λ is the wavelength of laser radiation; and $H = 60\lambda$.

Fig. 3 shows the angular spectra of the amplitude of the response field at the frequencies of the second and third harmonics for (s, s), (s, p), and (p, p) polarisations of the incident field. The propagation direction determined by Eqn (11) and the phase-matching condition (10) for each of the spectral components of the angular spectrum depend on the angle between the wave vectors of the waves and the refractive indices of the medium at the relevant frequencies. The sign of the parameter p corresponds to the sign of the projection of the wave vector of the response field on the y-axis.

In the case of an s, s polarised incident field, the response of the medium at the frequency of the second harmonic is p-polarised, while the polarisation of the field at the frequency of the third harmonic coincides with the polarisation of the incident field. The spectral component of the field response

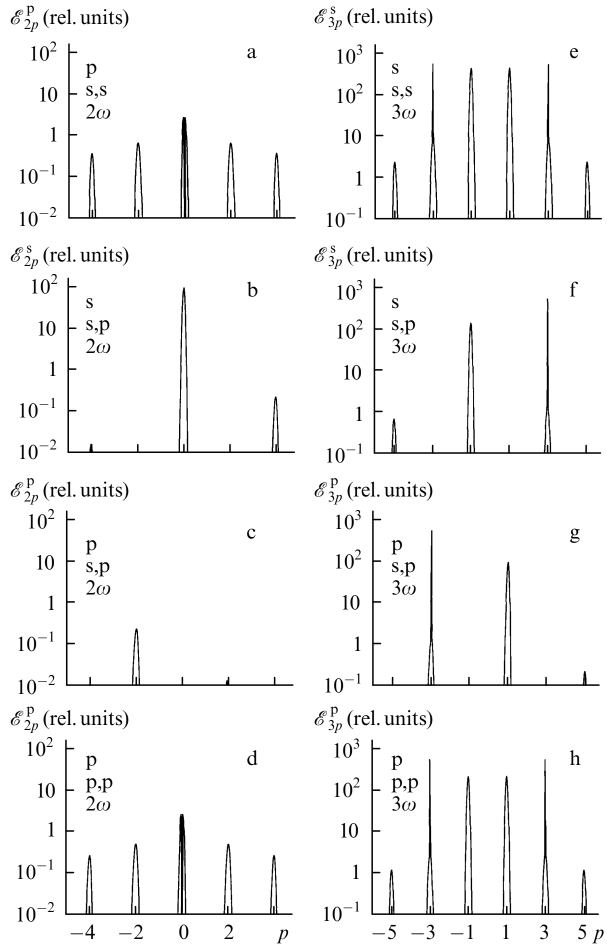

Figure 3. Angular spectra of the amplitudes of the response field for s (b) and p components (a, c, d) of the second harmonic and s (e, f) and p components (g, h) of the third harmonic for different polarisations of the external field: s,s (a, e), s,p (b, c, f, g), and p,p (d, h) polarisations.

at the frequency of the second harmonic ($p = 0$, Figs. 3a, 3d) has a double-humped structure, which is determined by the derivative of the profile of the incident field in the transverse coordinate. Second-harmonic radiation corresponding to this component propagates at a small angle to the z axis. This angle is inversely proportional to the pulse duration. The spectral components with $p = \pm 2$ (Figs. 3a–3d) correspond to second-harmonic generation in the direction coinciding with the propagation direction of incident pulses.

The second harmonic is generated due to a noncollinear interaction of two waves. Therefore, the angular spectrum of the second harmonic involves only broad spectral components, whose widths are inversely proportional to the transverse size S of the area where the wave packets overlap. Along with broad spectral components, the angular spectrum of the response field at the frequency of the third harmonic contains also narrow spectral components with $p = \pm 3$, which correspond to third-harmonic generation in the direction coinciding with the propagation direction of incident pulses. The widths of these spectral components are inversely proportional to the transverse size H of the medium, since each of these pulses independently generates the second harmonic not only in the area where the wave packets overlap, but also in the entire volume of the medium of two-level atoms.

In the case of an s, p-polarised incident field, no interference field is produced by the incident pulses. The response of the medium in this case is asymmetric with respect to the z-axis (Figs. 3b, 3c, 3f, 3g). The angular spectrum of radiation at the frequencies of the second and third harmonics involves components of both polarisations, which, however, emerge from the medium at different angles. The highest efficiency of SHG is achieved along the bisectrix of the angle between the wave vectors of the incident pulses ($p = 0$). In agreement with predictions of perturbation theory, the second harmonic generated in this direction is s polarised.

Polarisation of third-harmonic radiation propagating in the direction corresponding to $p = \pm 1$ (Figs. 3f, 3g) agrees well with the predictions of perturbation theory. An s polarised pulse propagating in the direction $p = +3$ generates the third harmonic propagating in the same direction and possessing the same polarisation. This result is confirmed by numerical simulations (Fig. 3f). A similar statement is also true for a p-polarised pulse of the field propagating in the direction $p = -3$ (Fig. 3g).

In the case of p,p-polarised field, the response field at the frequencies of the second and third harmonics is p-polarised. The s component of the atomic response is equal to zero in this case. We have additionally studied the dependences of the response field on the pulse duration and the amplitude of the external field in the case of a p,p-polarised incident field. As is clear from general physical analysis, the amplitudes of broad spectral components are proportional to the volume of the region where the wave packets overlap. If $S < H$, then this volume is proportional to the pulse duration squared.

However, not all the spectral components display these features. The spectral component with $p = 0$ (Fig. 3d) has a double-humped structure, which is determined by the derivative of the envelope of the external field. As a consequence, the amplitude of the spectral component with $p = 0$ is a linear function of the pulse duration. Narrow spectral components with $p = \pm 3$ (Fig. 3h) are produced in the entire volume of the medium of two-level atoms. Therefore, the amplitudes of these components are linear functions of the pulse duration.

Numerical simulations confirm the results of this qualitative analysis. The amplitudes of the spectral components of the second harmonic with $p = \pm 2$ and $p = \pm 4$ are quartic functions of the amplitude of the external field. In other words, these components are generated due to the fourth-order nonlinearity. The spectral component with $p = 0$ is generated due to the second-order nonlinearity. The spectral components of the third harmonic with $p = \pm 1$ and $p = \pm 3$ are cubic functions of the amplitude of the external field, while the components with $p = \pm 5$ increase with the fifth power of the amplitude of the external field.

Consider now the main results of numerical simulations for the cases when one of the pulses is circularly polarised or both pulses are circularly polarised. If the first pulse is s- or p-polarised and the second pulse is circularly polarised, then, in agreement with predictions of perturbation theory, the most intense signal at the frequency of the second harmonic is s polarised and propagates along the bisectrix of the angle between the wave vectors of incident pulses.

In contrast to the cases considered above, the second and third harmonics are generally elliptically polarised. In particular, polarisation of third-harmonic radiation with $p = -1$ is close to the circular one. The spectrum of the response at the frequency of the third harmonic features only one narrow spectral component with $p = +3$, which corresponds to the third harmonic generated by a linearly polarised field pulse in the entire volume of the medium of two-level atoms.

Third-harmonic generation with a circularly polarised pulse is characterised by a low efficiency. Therefore, the narrow spectral component with $p = -3$, corresponding to the emission of the third harmonic in the direction of propagation of the circularly polarised pulse, is not observed in this case. The response field at the frequency of the second harmonic is an order of magnitude weaker than the field at the frequency of the third harmonic.

When two circularly polarised pulses interact with a medium, we can distinguish between two cases. In the first case, both pulses have either right- or left-hand circular polarisation. In the second case, one of the pulses has a right-hand polarisation, while the other pulse has a left-hand circular polarisation. In agreement with predictions of perturbation theory, the highest efficiency of SHG is achieved in the latter case. Each of the circularly polarised pulses generates the third harmonic with a very low efficiency. Therefore, narrow spectral components are not observed in the angular spectrum of the third harmonic.

The most intense spectral components of the third harmonic with $p = \pm 1$ correspond to radiation propagating at an angle $\varphi_{31} \approx 11^\circ$ with respect to the z-axis. Polarisation of this radiation is close to the circular polarisation. In the latter case, the amplitudes of the second and third harmonics are of the same order of magnitude. We should note that the angular spectrum of the response field in both cases is symmetric with respect to the longitudinal z-axis.

6. Conclusions

Thus, the developed theory of interaction of atoms with a superstrong spatially inhomogeneous laser field describes, in the two-level approximation, the dynamics of spatially nonlocal interactions determined by variations in level populations. The proposed model includes spatially nonlocal interactions (magnetic-dipole and quadrupole interactions, as well as interactions due to the gradient of the ponderomotive potential of the field) with an accuracy up to the first spatial derivative of the vector potential of the field. The developed model can be employed to analyse both resonant and nonresonant interactions of an atom with ultrashort pulses of the field of arbitrary durations.

Comparison of the results of numerical simulations with the predictions of stationary perturbation theory has shown that, within the framework of stationary perturbation theory, the second harmonic is generated only along the bisectrix of the angle between the wave vectors of the waves interacting with the medium. The response field at the frequency of the second harmonic is s-polarised, and the amplitude of this field depends on the polarisation of the incident field. The inclusion of the dynamics of populations in atomic levels and a finite pulse duration gives rise to new spectral components in the angular spectrum of the response field.

We have investigated the specific features of the nonlinear-optical response of an atom in a strong field. As the field amplitude grows, variations of level populations increase the refractive index of an initially nonexcited medium if the field frequency exceeds the frequency of the atomic transition. In the opposite case, the refractive index of the medium decreases. The atomic response at the frequencies of the second and third harmonics saturates with the growth in the amplitude of the incident field.

We have performed numerical simulations for the angular spectra of the response field of the second and third harmonics for different polarisations of the external field. We have also derived analytic expressions for the phase-matching condition and the dependence of the angular distribution of the response field on the angle between the wave vectors of the interacting waves and the refractive indices of the medium at the relevant frequencies.

Acknowledgements. This study was partially supported by the Russian Foundation for Basic Research (Grant No. 99-02-16093), 'Fundamental Optics and Spectroscopy' Education and Research Centre, and the 'Universities of Russia' program.

References

1. Platonenko V T, Strelkov V V *Kvantovaya Elektron.* **25** 582 (1998) [*Quantum Electron.* **28** 564 (1998)]
2. Kim A V, Ryabikin M Yu, Sergeev A M *Usp. Fiz. Nauk* **169** 58 (1999) [*Phys. Usp.* **42** 54 (1999)]
3. Babin A A, Kiselev A M, Pravdenko K I, Sergeev A M, Stepanov A N, Khazanov E A *Usp. Fiz. Nauk* **169** 80 (1999) [*Phys. Usp.* **42** 74 (1999)]
4. Fedorov M V *Usp. Fiz. Nauk* **169** 66 (1999) [*Phys. Usp.* **42** 61 (1999)]
5. Andreev A V, Kozlov A B *Proc. SPIE Int. Soc. Opt. Eng.* **3735** 75 (1999)
6. Nisoli M, De Silvestri S, Svelto O, Szipocs R, Ferencz K, Spielmann Ch, Sartania S, Krausz F *Opt. Lett.* **22** 522 (1997)
7. Akhmanov S A, Khokhlov R V *Problemy Nelineinoi Optiki* (Problems of Nonlinear Optics) (Moscow: VINITI, 1964)
8. Kleinman D A *Phys. Rev.* **126** 1977 (1962)
9. Bethune D S *Phys. Rev. A* **23**, 3139 (1981)
10. Andreev A V, Andreeva O A, Balakin A V, Boucher D, Masselin P, Ozheredov I A, Prudnikov I R, Shkurinov A P *Kvantovaya Elektron.* **28** 75 (1999) [*Quantum Electron.* **29** 632 (1999)]
11. Shen Y R *The Principles of Nonlinear Optics* (New York: Wiley, 1984)
12. Mossberg T, Flusberg A, Hartmann S R *Opt. Commun.* **25** 121 (1978)
13. Kim D, Mullin C S, Shen Y R *J. Opt. Soc. Am. B* **14** 2530 (1997)
14. Miyazaki K, Sato T, Kashiwagi H *Phys. Rev. Lett.* **43** 1154 (1979)
15. Malcuit M S, Boyd R W, Davis W V, Rzazewski K *Phys. Rev. A* **41** 3822 (1990)
16. Dinev S *J. Phys. B* **21** 1681 (1988)
17. Jamroz W, LaRocque P E, Stoicheff B P *Opt. Lett.* **7** 148 (1982)
18. Hakuta K, Marmet L, Stoicheff B P *Phys. Rev. Lett.* **66** 596 (1991)
19. Bethune D S, Smith R W, Shen Y R *Phys. Rev. A* **17** 277 (1978)
20. Andreev A V *Zh. Eksp. Teor. Fiz.* **116** 793 (1999)