

# Polarisation response of a gas medium in the field of a high-intensity ultrashort laser pulse: high order Kerr nonlinearities or plasma electron component?

E.A. Volkova, A.M. Popov, O.V. Tikhonova

**Abstract.** The polarisation response of quantum systems modelling silver and xenon atoms in the field of a high-intensity femtosecond Ti:sapphire laser (photon energy  $\hbar\omega \approx 1.5$  eV), has been investigated by direct numerical integration of the Schrödinger equation. The applicability ranges of the perturbation theory and polarisation expansion in powers of field are determined. The contributions of excited atoms and electrons in the continuous-spectrum states to the polarisation response at the fundamental frequency, which arise as a result of excitation and photoionisation, are analysed. It is shown that specifically ionisation changes the sign of dielectric susceptibility with an increase in radiation intensity for the systems under consideration.

**Keywords:** self-focusing, polarisation response, multiphoton ionisation, numerical simulation, strong electromagnetic fields.

## 1. Introduction

The effect of self-focusing of electromagnetic radiation in nonlinear media, which was predicted 50 years ago [1], considered in the theoretical studies [2, 3], and experimentally observed for the first time in [4], has been in focus of researchers' attention for several decades. A conventional concept of self-focusing is as follows: application of a sufficiently strong laser field to a medium results in nonlinear polarisability at the fundamental frequency in this medium, which leads to the formation of a collecting lens to focus the propagating radiation. If the field strength in the electromagnetic wave is small in comparison with the intra-atomic field, nonlinear polarisabilities can be calculated within the quantum-mechanical perturbation theory. A term linear in field (which is used in linear optics) arises in the first order of the perturbation theory, a quadratic term arises in the second order, the third order yields a cubic term, etc. The contribution to the polarisation response on the fundamental frequency yields all nonlinear susceptibilities of odd orders. However, within the above-described approach, the main contribution to the nonlinear response is from the cubic susceptibility of the medium.

The additional (small) contribution to the nonlinear polarisability at the fundamental frequency yields also nonlinear susceptibilities of the fifth, seventh, and higher odd orders. In other words, within the above-described approach the atomic response at the fundamental frequency can be written as a series:

$$d_{\omega} = \chi(\omega, E_{\omega}) E_{\omega} = \chi^{(1)}(\omega) E_{\omega} + \chi^{(3)}(\omega) E_{\omega}^3 + \dots, \quad (1)$$

where  $\chi^{(1)}(\omega)$  and  $\chi^{(3)}(\omega)$  are, respectively, the linear and cubic nonlinear susceptibilities of the medium at the field frequency.

Here, the limiting size of the spot into which a laser beam can be focused is determined by the free-electron density in the plasma formed during beam compression. The reason is that plasma has negative susceptibility, due to which an increase in the electron concentration is accompanied by compensation of the focusing properties of the medium. As a result, radiation self-focusing generally leads to the formation of one or several long conducting channels (filaments); they are nonequilibrium plasma regions emitting in a wide frequency range [5, 6].

The development of high-power terawatt lasers, which can generate pulses with a wave electric field strength comparable with the intra-atomic values (or even higher) and widths of several tens of femtoseconds, makes urgent study of self-focusing in this new range of laser parameters (see [7, 8] and references therein). Under these conditions, the quantum-mechanical perturbation theory is invalid, and, therefore, the expansion in powers of field for calculating atomic susceptibilities is incorrect. At the same time, there are hardly any collisions between gas particles during pulses of several tens of femtoseconds, and the plasma electron component is formed only as a result of multiphoton or tunnel (above-barrier) ionisation of atoms, whereas avalanche ionisation can be neglected in this case.

Actually, in this situation the polarisation response of the medium can be calculated by studying the dynamics of electrons of one of the atoms of the medium in the field of a high-intensity ultrashort pulse. Currently this problem can be numerically solved by direct integration of the time-dependent Schrödinger equation, which describes the evolution of electrons of an atom (molecule) in an electromagnetic wave field [9]. This approach is now very urgent. It was suggested in [10–12] that, along with plasma formation, nonlinearities due to the high-order Kerr effect (HOKE) can also play an important role in self-focusing and filamentation. Indeed, if high-

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Received 28 April 2012; revision received 14 June 2012  
Kvantovaya Elektronika 42 (8) 680–686 (2012)  
Translated by Yu.P. Sin'kov

order nonlinearities make a negative contribution to the refractive index, limitation of the size of laser beam compression as a result of filamentation may be theoretically possible even in the absence of ionisation under certain conditions. This concept was recently confirmed by the experimental studies [13, 14], where an attempt was made to explain the filamentation of laser radiation with intensities of several tens of TW cm<sup>-2</sup> by the dominant contribution or higher orders nonlinearities to the response when plasma is not formed. However, this interpretation of the experiments [13, 14] was doubted in [15, 16].

A number of publications have appeared in the last two years, both supporting the new point of view on the role of HOKE in the laser beam filamentation (see, for example, [13, 14, 17–20]) and disproving it (see [15, 16, 21–25]). Without going into the details of the discussion between the adherents and antagonists of HOKE, we should admit that there are no convincing experimental data in favour of a particular point of view. In our opinion, the role of the plasma electron component and the nonlinearities due to neutral atoms in the filamentation of ultrashort laser pulses can be unambiguously determined by direct numerical solution of the time-dependent Schrödinger equation, which describes the dynamics of the electron subsystem of atom in laser field and makes it possible to distinguish the effects related to the atomic polarisation and the formation of free electrons. Specifically this is the problem of our study.

## 2. Statement of the problem

We performed numerical calculations for single-electron atoms with the ionisation potentials  $I = 7.58$  and  $12.13$  eV, which correspond to those of silver and xenon atoms. The single-electron model of a silver atom was discussed in detail in [26]. The quantum system with an ionisation potential of  $12.13$  eV, corresponding to the xenon atom, was described by the smoothed Coulomb potential

$$V(r) = -\frac{1}{\sqrt{\alpha^2 + r^2}}$$

with the smoothing parameter  $\alpha = 0.0943$  Å. According to the calculations, this model describes well the dependence of the ionisation probability of xenon atom on the radiation intensity and is in agreement with the experimental data of [27, 28].

The evolution of the electron subsystem of atom in an external electromagnetic field was analysed based on numerical solution of the time-dependent Schrödinger equation:

$$i\frac{\partial\psi(\mathbf{r}, t)}{\partial t} = -\frac{1}{2}\nabla^2\psi(\mathbf{r}, t) + V(r)\psi(\mathbf{r}, t) + V_{\text{int}}(\mathbf{r}, t)\psi(\mathbf{r}, t). \quad (2)$$

Here,  $V(r)$  is the single-electron potential of the atom studied and  $V_{\text{int}}(\mathbf{r}, t)$  is the interaction energy of this atom with the electromagnetic wave. The electromagnetic wave field was assumed to be linearly polarised, and its interaction with the atom was analysed in the dipole approximation:

$$V_{\text{int}} = r \cos\theta E(t), \quad (3)$$

where  $E(t)$  is the wave electric field strength and  $\theta$  is the angle between the electron radius vector  $\mathbf{r}$  and the vector  $\mathbf{E}(t)$ . It

was assumed that at the initial instant the atom is in the ground state  $|g\rangle$ . The laser pulse had a smoothed trapezoidal shape with front and plateau durations  $t_f$  and  $t_p$ , respectively. The electric field was set in terms of the vector potential  $E(t) = -(1/c)/(\partial A/\partial t)$

$$A(t) = A_0(t) \sin \omega t,$$

with the envelope

$$A_0(t) = \begin{cases} A_0 \sin^2 \frac{\pi t}{2t_f}, & t \leq t_f, \\ A_0, & t_f \leq t \leq t_f + t_p, \\ A_0 \cos^2 \frac{\pi[t - (t_f + t_p)]}{2t_f}, & t_f + t_p \leq t \leq 2t_f + t_p. \end{cases} \quad (4)$$

The amplitude of the vector potential is related to the radiation intensity in the pulse plateau region as follows:  $P = \omega^2 A_0^2 / 8\pi c$ .

The calculations were performed with the photon energy  $\hbar\omega = 1.5$  eV, a value approximately corresponding to the lasing frequency of the Ti:sapphire laser. Two different laser pulse shapes were used: a ‘short’ pulse with  $t_f = 2T$  and  $t_p = 10T$  ( $T = 2\pi/\omega$  is the optical oscillation period) for calculating the evolution of silver atom and a ‘long’ pulse with  $t_f = 20T$  and  $t_p = 0$  for the xenon-like system.

The technique of numerical integration of Eqn (2) was discussed in detail in [29]. The polarisation response of an atomic system can be calculated from the wave function of the system obtained as a result of integration of Eqn (2):

$$\langle d_z(t) \rangle = -\int r \cos(\theta) |\psi(\mathbf{r}, t)|^2 d^3r. \quad (5)$$

Note that the dipole moment arising in the case under consideration is directed along the radiation polarisation vector.

Expansion of the functions  $E(t)$  and  $\langle d_z(t) \rangle$  in the Fourier integral,

$$E_\Omega = \frac{1}{\sqrt{2\pi}} \int E(t) \exp(-i\Omega t) dt, \quad (6)$$

$$d_\Omega = \frac{1}{\sqrt{2\pi}} \int \langle d_z(t) \rangle \exp(-i\Omega t) dt, \quad (7)$$

makes it possible to determine the spectral composition of the polarisation response  $d_\Omega(E_\Omega)$  and perform *ab initio* calculation of the nonlinear susceptibilities of different orders, find the range of application for the calculation of response by expansion in field powers, and reveal the contribution of neutral atoms and free electrons to the polarisation response.

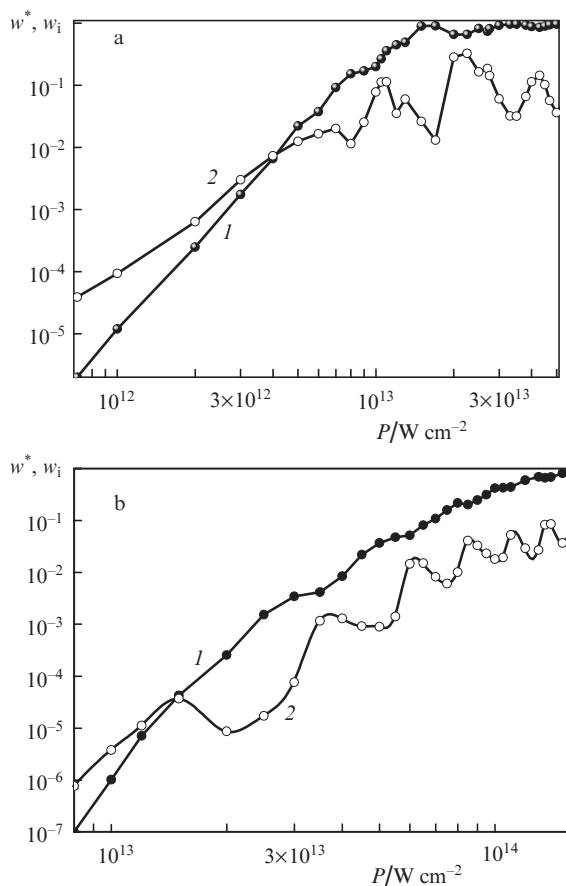
In practice, it is more convenient to carry out the corresponding calculations using the Ehrenfest theorem [30]

$$\langle a_z(t) \rangle = -E(t) - \left\langle \frac{\partial V}{\partial z} \right\rangle, \quad (8)$$

because the Fourier transforms of the dipole moment  $d_\Omega$  and the acceleration  $a_\Omega$  are related by the expression  $d_\Omega = a_\Omega / \Omega^2$ .

### 3. Simulation results

Let us primarily analyse the dependence of the calculated ionisation and excitation probabilities for the atoms under study on the Ti:sapphire laser radiation intensity. The calculated ionisation probabilities  $w_i(P)$  and excitation\* probabilities  $w^*(P)$  of atomic states (Fig. 1) by the end of laser irradiation for weak fields correspond to the lowest order of the perturbation theory. For stronger fields ( $P \geq 10^{13} \text{ W cm}^{-2}$  for the silver atom and  $P \geq 3 \times 10^{13} \text{ W cm}^{-2}$  for the xenon-like system), the dependences obtained are nonmonotonic, which is especially pronounced in the curve  $w^*(P)$ . The probability of finding an atom in the excited state turned out to be an oscillating function of intensity. It was shown in [26, 28] that the effect observed is based on the multiphoton resonance between the ground state and the group of excited (Rydberg) states, which leads to their population and subsequent coherent repopulation via Raman transitions of the  $\Lambda$  type.



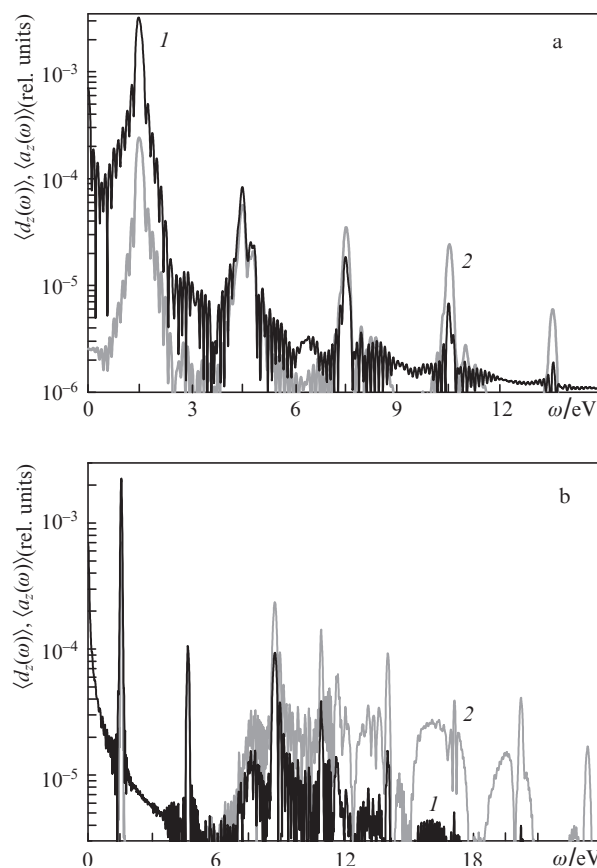
**Figure 1.** Dependences of the (1) ionisation and (2) excitation probabilities of (a) silver and (b) xenon atoms irradiated by a Ti:sapphire laser pulse on the laser radiation intensity.

Obviously, the polarisation response of the system in the range of intensities corresponding to onset of trapping atoms in the excited (primarily, Rydberg) states, cannot be calculated within the method of polarisation expansion in powers of field, in particular, with allowance for HOKE. As will be

\*) The excitation probability is considered to be the total probability for population of all bound states, except for the initial (ground) one.

shown below, trapping of atoms in Rydberg states significantly affects the nonlinear atomic susceptibilities.

To analyse the dependence of the nonlinear atomic response on the electric wave field for different laser intensities from the calculated wave function of the system,  $\psi(\mathbf{r}, t)$ , and using (5) and (8), we obtained the functions  $\langle d_z(t) \rangle$  and  $\langle a_z(t) \rangle$  and then expanded them in the Fourier integral. The typical results of the spectral expansion of these functions for the quantum systems under study are shown in Fig. 2. These expansions have pronounced peaks at the frequencies corresponding to odd harmonics of the laser pulse frequency  $\Omega_{2n+1} = (2n+1)\omega$  ( $n = 0, 1, 2, \dots$ ). Hence, one can calculate *ab initio* the arising polarisations at the frequencies  $\Omega_{2n+1}$ , taking into account the ionisation and excitation of different atomic states; determine the intensity range in which the polarisation response can be expanded in powers of field; and investigate the contribution of the atomic ionisation and excitation to the polarisation response at the fundamental frequency.

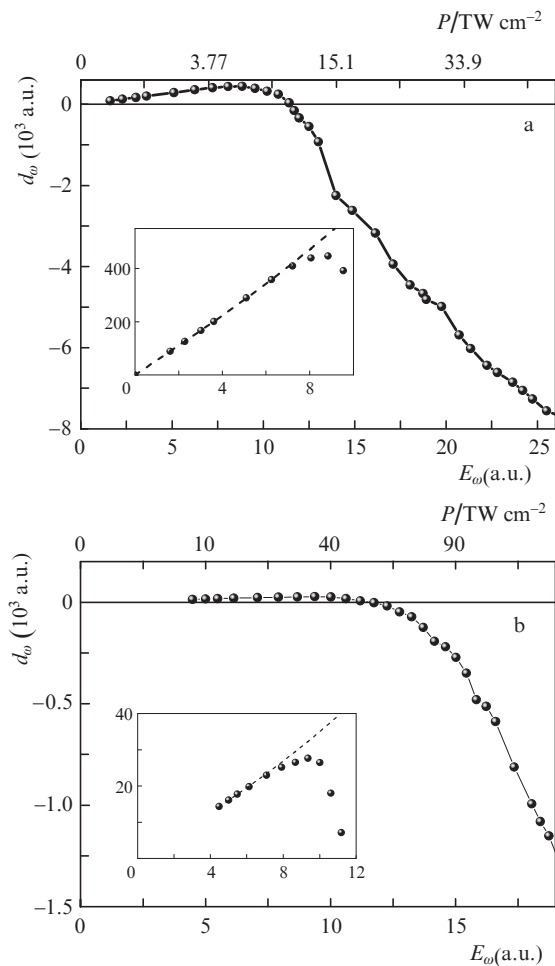


**Figure 2.** Spectral expansions of the (1) dipole moment  $\langle d_z(t) \rangle$  and (2) the electron acceleration  $\langle a_z(t) \rangle$ , calculated (a) for silver atoms at an intensity of  $1.5 \times 10^{13} \text{ W cm}^{-2}$  and (b) for xenon atoms at an intensity of  $4 \times 10^{13} \text{ W cm}^{-2}$ .

The calculated dependences of the polarisation response at the laser field frequency  $d_\omega$  on the spectral strength of the wave electric field for both atoms are shown in Fig. 3\*).

\*) One should take into account that the pulses used in the calculations have different durations and, therefore, different spectral widths. Hence, the same value of peak radiation intensity corresponds to different values of the spectral electric field strength  $E_\omega$  for pulses of different durations.

relatively weak laser fields, at which an irradiated atom remains generally in the ground state, the obtained dependences are indeed described by an expression of type (1). However, when the intensity exceeds some critical value, characteristic of a specific atomic system, one first observes saturation of the polarisation response, after which it changes sign and becomes negative. In fact this means that, when the radiation intensity increases, the focusing properties of the medium turn to defocusing, giving rise to filamentation.



**Figure 3.** Dependences of the polarisation response at the field frequency on the spectral amplitude of the wave electric field strength (or on the radiation intensity) for (a) silver and (b) xenon atoms. The insets show the same dependences for weak fields, which make it possible to determine the range of application of polarisation expansion in powers of field. The dashed lines show dependence (1).

The obtained dependence  $d_\omega(E_\omega)$  can be due to the contribution of higher order nonlinearities in (1), as was suggested in [13, 14, 17–20]. At the same time, the saturation of the polarisation response of the system with an increase in the field and its subsequent decrease with a change in the sign of  $d_\omega$  can also be caused by both the contribution of excited atomic states to the polarisation response and the photoionisation and formation of the plasma electron component. As was noted above, in the case of extremely short pulses, the collisional processes in the gas can be neglected, and the polarisation response of the medium is determined in fact by the response of an individual atom. Specifically this circum-

stance makes it possible to analyse (based on numerical solution of the Schrödinger equation for an atomic system in the electromagnetic wave field) the contributions of different processes, including excitation of different atomic states and photoionisation atoms, to the response.

Indeed, the susceptibility of a free electron gas (collisionless plasma), recalculated per electron, is determined by the expression  $\chi_e(\omega) = -1/\omega^2$ . This value for a photon of Ti:sapphire laser radiation is about an order of magnitude larger than the linear atomic susceptibilities  $\chi^{(1)}(\omega)$  of xenon and silver atoms.

Therefore, even at a degree of gas ionisation of about 10%, the contribution of free electrons in the continuum to the polarisability of the medium should be significant. Specifically this situation is observed in our calculations: for example, for the model xenon atom at a radiation intensity of  $\sim 5 \times 10^{13} \text{ W cm}^{-2}$  (a value at which  $d_\omega$  becomes zero) the ionisation probability of atoms is 3%–5%. Similarly,  $d_\omega$  changes sign for silver atoms at an intensity of  $10^{13} \text{ W cm}^{-2}$ , when the ionisation probability is  $\sim 10\%$ .

However, it should be noted that quantitative analysis of the contribution of different atomic states to the polarisation response in strong laser fields meets a number of difficulties. The most fundamental one is that the population of specific atomic states during laser irradiation in the nonperturbative ionisation regime can be considered in a rather conditional sense, on the assumption of a specific basis of atomic states of the discrete and continuous spectra. Generally speaking, it is incorrect to use the basis of free atomic states in this case, because exposure to a strong radiation field leads to a significant reconstruction of the atomic states and formation of field-dressed atom. Thus, the contribution of different atomic states (including the states of continuum) to the response can be calculated only within a specific model, describing the structure of the dressed-atom spectrum and the temporal dynamics of the populations of the field-reconstructed states.

There is another important circumstance. The concept of  $n$ th-order atomic susceptibility is introduced within the quantum-mechanical perturbation theory on the assumption that the population amplitudes for all states (including the states of the discrete spectrum and continuum) are small in comparison with the population amplitude of the initial (generally, ground) state. Therefore, even on the assumption that the atomic response can be analysed in the basis of unperturbed-atom states, in the case of strong laser fields (where the wave function can be written as a coherent superposition of a large number of stationary atomic states of the discrete spectrum and continuum with comparable amplitudes), the description of the response of the system in terms of different-order susceptibilities is, generally speaking, impossible.

An important fact for further discussion concerning interpretation of the data obtained is that a rather large part of the residual population of bound states corresponds to highly excited Rydberg states near the continuum boundary in the case of strong laser fields. We should primarily note that, at interference stabilisation, Rydberg states are populated at the leading edge of generation, and this population is retained at a constant level (see [26]), whereas the probability of transition of an electron in the continuum monotonically increases during the laser pulse. When analysing the response at the field frequency, this circumstance makes it possible to exclude the interference terms containing the population amplitudes for the ground and low-lying excited atomic states. Since the

polarisability of Rydberg states almost coincides with that of free electrons  $\chi_e(\omega) = -1/\omega^2$  [31], the contribution of excited (Rydberg) states to the atomic response is  $d_{\omega}^{(\text{Rydb})} = -w^{(\text{Rydb})} E_{\omega}/\omega^2$ , where  $w^{(\text{Rydb})}$  is the excitation probability for Rydberg atomic states. Now we also take into account the presence of neutral atoms in the ground state, the susceptibility of which can be calculated using expansion in powers of field:

$$\chi_g(\omega, E_{\omega}) = \chi_g^{(1)}(\omega) + \chi_g^{(3)}(\omega) E_{\omega}^2$$

[here,  $\chi_g^{(1)}(\omega)$  and  $\chi_g^{(3)}(\omega)$  are, respectively, the linear and cubic susceptibilities of neutral atoms at the field frequency]. The higher terms in the expansion of susceptibility in powers of field are disregarded, because the concentration of neutral atoms in strong fields is negligible, and the form of the function  $\chi_g(\omega, E_{\omega})$  is of little importance. With allowance for the aforesaid, we can estimate the contribution of free electrons to the susceptibility of the medium:

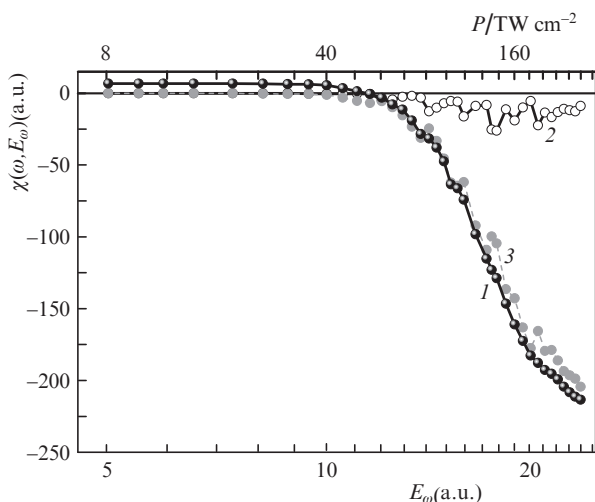
$$\chi_e(\omega, E_{\omega}) = d_{\omega}(E_{\omega})/E_{\omega} + w^{(\text{Rydb})}(E_{\omega})/\omega^2$$

$$-w_g(E_{\omega})\chi_g(\omega, E_{\omega}), \quad (9)$$

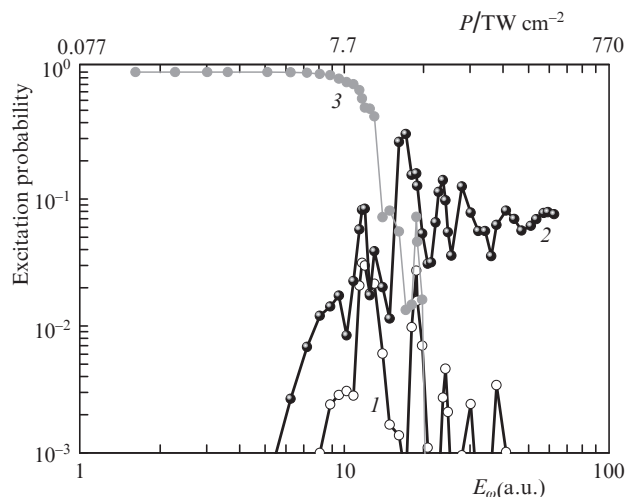
where  $w_g$  is the probability of detecting an atom in the ground state by the end of the pulse.

The thus calculated contributions of free electrons and Rydberg atoms to the susceptibility of model xenon atoms is shown in Fig. 4. It can be seen that free electrons make a dominant contribution to the nonlinear susceptibility at intensities  $P \geq 5 \times 10^{13} \text{ W cm}^{-2}$ ; i.e., within the model under consideration, radiation is filamented as a result of gas photoionisation and plasma formation. It can also be seen that the stabilisation and trapping of some part of population in Rydberg states also contributes to filamentation.

The situation where the contributions of different states to the susceptibility of silver atom in the strong field of Ti:sap-



**Figure 4.** Dependences of the (1) susceptibility of model xenon atoms,  $\chi(\omega, E_{\omega}) = d_{\omega}(E_{\omega})/E_{\omega}$ , at the field frequency and (2, 3) the contributions of (2) Rydberg atoms and (3) free electrons to the susceptibility on the spectral amplitude of the wave electric field (on the radiation intensity).

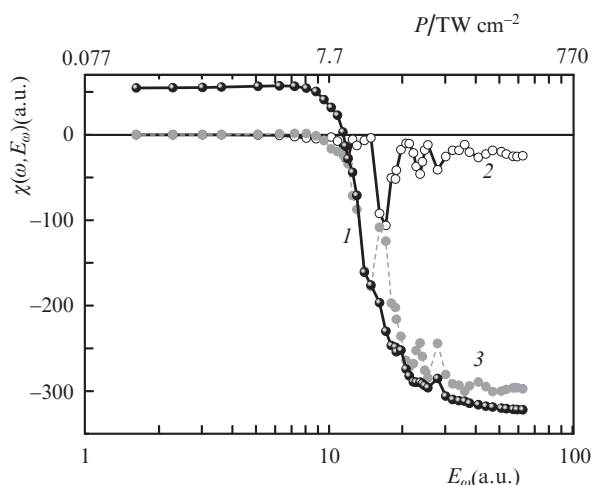


**Figure 5.** Dependences of the excitation probabilities of (1) low-lying and (2) Rydberg states of the silver atom on the spectral amplitude of the wave electric field (on the radiation intensity). Curve (3) is the probability of finding the atom in the ground state.

phire laser radiation are analysed is more complex. The presence of low-lying<sup>\*</sup> excited states leads to a rather high probability of their population as a result of three- and four-photon absorption if the difference in the ground- and excited-state energies (with allowance for their Stark shift) coincides with the sum energy of an integer number of field photons. The dependences of the excitation probability for low-lying and Rydberg states of discrete spectrum on the intensity (and the spectral amplitude of the wave electric field strength) are shown in Fig. 5. It can be seen that the probability of exciting Rydberg states dominates practically at all radiation intensities. The most pronounced multiphoton resonance, which is due to the three-photon excitation of the lower p state, is observed at intensities close to  $\sim 10^{13} \text{ W cm}^{-2}$ .

Specifically the necessity of taking into account the contribution of low-lying excited states is, generally speaking, an additional problem. Indeed, near the resonance their susceptibility can take both positive and negative values; note also that the susceptibility of an excited atom may exceed significantly that of an atom in the ground state. Therefore, having subtracted the contributions of Rydberg atoms and the ground atomic state [see expression (9)] from the atomic susceptibility found in numerical calculations, we obtain the sum of susceptibilities of the electrons that are in the continuum and in the low-lying excited atomic states  $\tilde{\chi}(\omega, E_{\omega})$  [see Fig. 6, curve (3)]. However, in our calculations, the contribution of low-lying excited states is small at any radiation intensities. This conclusion stems from the fact that the dependence  $\tilde{\chi}(\omega, E_{\omega})$  exhibits no features in the intensity range corresponding to the resonant population of low-lying states  $[(1-2) \times 10^{13} \text{ W cm}^{-2}]$ . At the same time, the positions of maxima and minima in this dependence exactly coincide with the positions of maxima and minima in the curve describing the ionisation probability. Therefore, one can state that, under the conditions analysed here, the contribution of low-lying excited states to the susceptibility is insignificant, and  $\tilde{\chi}(\omega, E_{\omega})$  describes in fact the contribution of the plasma electron com-

<sup>\*</sup> Here, low-lying excited states are considered to be the ones bound to the continuum through multiphoton transitions.



**Figure 6.** Dependences of the (1) susceptibility of model silver atoms,  $\chi(\omega, E_\omega) = d_\omega(E_\omega)/E_\omega$ , at the field frequency and (2, 3) the contributions of (2) Rydberg atoms and (3) free electrons to the susceptibility on the spectral amplitude of the wave electric field (on the radiation intensity).

ponent. Therefore, the saturation of the response and the change in its sign for both silver and xenon atoms are also caused by the photoionisation and stabilisation of highly excited atomic states.

The above data convincingly show that, under the conditions considered in our calculations, the saturation of the response and the change in its sign are primarily caused by the plasma formation. At the same time, in strong fields (case where the atomic dynamics is nonperturbative), as can be seen in Figs 4 and 6, the response of neutral, highly excited atoms also play an important role. The susceptibility of Rydberg atoms is also negative (i.e., it enhances the defocusing properties of the medium). The susceptibilities of Rydberg atoms and free electrons almost coincide, as a result of which it is difficult to measure their contributions to the response saturation and filamentation. Apparently, a possible way to implement propagation of a high-power laser pulse and its filamentation is to accumulate Rydberg atoms in the medium in the absence of significant contribution from free electrons.

#### 4. Conclusions

Thus, the numerical calculations of the polarisation response of model silver and xenon atoms in the field of a high-intensity femtosecond laser pulse showed limitations of the generally used approach, which is based on the response expansion in powers of electric field and introduction of nonlinear susceptibilities of different orders. At radiation intensities exceeding  $10^{13} \text{ W cm}^{-2}$  (which is approximately three orders of magnitude smaller than the atomic value of intensity), the electron dynamics in atoms is significantly nonperturbative and cannot be described within the quantum-mechanical perturbation theory, which is the basis of the aforementioned method. In particular, in fields corresponding to the nonperturbative regime, the most important effect taken into account in calculation of the response at the frequency of wave actuating field is the nonlinear atomic ionisation, which leads to plasma formation and is responsible for the filamentation of high-power ultrashort laser pulses in different media.

Formation of Rydberg atoms, stable with respect to ionisation, also contributes to the nonlinear response and facilitates laser beam filamentation. However, there are no grounds to revise the existing paradigm of filamentation of ultrashort laser pulses (as this was done in [13, 14, 17–20]) for the conditions of our calculation.

**Acknowledgements.** This work was supported by the Russian Foundation for Basic Research (Grant No. 12-02-00064) and the Ministry of Education and Science of the Russian Federation. Numerical integration of the Schrödinger equation was performed on the SKIF-MGU ‘Chebyshev’ supercomputer.

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