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A Variationally Stable Method in the Problem of Two-Photon Atomic Ionization

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Abstract—A variationally stable method is analyzed; it is one of the rarely used approaches for calculating the amplitudes of nonlinear photoprocesses in the framework of perturbation theory. The convergence of the method is studied based on the example of atomic hydrogen and hydrogenlike ions. The method is used to calculate the angular distributions of photoelectrons for the first time.

Keywords: photoionization of atoms and ions, nonlinear photoprocesses, multiphoton ionization, angular distribution of electrons, VUV and X-ray radiation, perturbation theory.

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INTRODUCTION

Multiphoton ionization is the process of ion formation as a result of absorption of several photons in one elementary act. This is one of the fundamental processes of nonlinear optics. For many years, the theoretical regularities and experimental aspects of this process have been an object of intense studies (see, for example, [1] and references). Until recently the phenomenon of multiphoton ionization was observed (with a few exceptions) under the action of optical or infrared laser fields, since there were no sources with sufficiently large intensity in the short-wavelength range of spectrum. The observation of nonlinear photoprocesses in the range of vacuum ultraviolet (VUV) and soft X-rays has become possible recently due to the use of X-ray lasers based on free electrons, whose intensity is many orders of magnitude higher than the intensity of synchrotron radiation of the preceding generations. As a result, a rapidly developing field has appeared that is related to the investigation of the multiphoton processes under the action of intense pulses of soft X-ray and VUV radiation [2–4]. The examples of such processes are single-photon and two-photon ionization from deep atomic shells, consecutive two-photon and triple-photon ionization, two-frequency ionization with changes in the relative polarization of photonic beams and other processes, whose study in the X-ray and VUV ranges of spectra was impossible until recently.

Regarding the theoretical research, it is necessary to note the following. In the case of radiation in the range from infrared to ultraviolet, i.e. for photon energies up to several eV, the intensities on the order of

10^{13} – 10^{15} W/cm² correspond to a strong field in which the processes of interaction cannot be described by perturbation theory, such as tunneling and generation of high harmonics. A criterion of the implementation of this regime is the ratio between the ponderomotive energy of an electron in a laser field and the photon energy. Since this ratio decreases with increasing photon energy as the photon frequency cubed, then in the range of VUV at the same intensities as in the experiments with X-ray lasers on free electrons, the lowest-order perturbation theory provides, as a rule, an adequate base for describing the processes. Thus, the high importance of the perturbation theory for nonlinear optics in the VUV and X-ray ranges follows. However, a strict approach to computing the amplitudes of perturbation theory already in the second order assumes the summation over an infinite set of intermediate states, including integration over a continuous spectrum. Such calculations are carried out by direct summation or using Green functions; these are rather complicated. The situation is aggravated upon transition to the perturbation theory of the third and higher orders. Therefore, elaborating alternative approaches to calculation of the amplitudes of photoprocesses within the perturbation theory is urgent.

In this work, our attention is focused on calculating the characteristics of the process of two-photon ionization within the second-order perturbation theory based on the variationally stable principle, which is a variationally stable method [5] that was first described in [6, 7]. This method is not related to an explicit summation over the intermediate states of the system in calculating the matrix elements of multiphoton transitions.

A variational procedure is used instead of such a summation. The method has been successfully applied several times, in the calculations of the total cross sections of multiphoton ionization of atoms, negative ions and molecules [6–12], static and dynamic polarizability of atoms, ions and molecules [11–18], and the constants of dispersion forces (van-der-Waals forces) [17–19]. The results for sevenfold ionization of the hydrogen atom that were obtained using the variationally stable method proved to be in excellent agreement with the results of direct summation over the intermediate states using the Green-function method [20] for a wide range of wavelengths of incident radiation. Therefore, various calculations indicate that the considered method has a great potential when solving problems of high-order perturbation theory. Despite this fact, the range of problems that have been solved by the variationally stable method is very narrow. This method has not been used in calculating the angular distributions of photoelectrons and other differential characteristics of photoionization, as well as the photoionization in two- and multifrequency fields. The method has been insufficiently tested in the calculations of two- and multiphoton ionization of multielectron atoms. Studies of the convergence of this method to an accurate solution by the perturbation theory and the criteria for choosing the class of effective states are also not found in the literature.

Extension of the practice of using variationally stable method requires a more detailed study of its features. In this work, the variationally stable method is implemented in the problem of two-photon ionization of the hydrogen atom and hydrogenlike ions in the range of VUV. Based on this simple example, by knowing the accurate atomic wave functions, we analyze the limits of the applicability of the method and its convergence to the accurate results of the second-order perturbation theory. We also use the variationally stable method for calculating the angular distributions of photoelectrons.

The first section of this work presents the basic statements of the method as applied to the process of two-photon ionization. We discuss a concrete way to perform the method and its advantages and disadvantages. In the second section, the total cross sections of the two-photon ionization of the ground atomic states of hydrogen and hydrogenlike ions are given calculated using the second-order perturbation theory and the variationally stable method. The choice of the parameters of trial functions of effective states is discussed. Comparison with the accurate results of the second-order perturbation theory is carried out to analyze the ranges of applicability of the variationally stable method. Furthermore, the method is applied in calculating the angular distributions of photoelectrons. In conclusion, we formulate the inferences from the research.

1. BASIC STATEMENTS OF THE VARIATIONALLY STABLE METHOD

We recall the main statements of the variationally stable method based on the example of two-photon ionization of a quantum system (atom) [9]. Within the second-order perturbation theory, the formula that describes the amplitude of a two-photon transition from state i of the discrete spectrum to state f of the continuous spectrum is given by [21]

$$T_{i \rightarrow f}^{(2)} = \sum_s \frac{\langle f | \hat{D} | s \rangle \langle s | \hat{D} | i \rangle}{E_i - E_s + \omega}, \quad (1)$$

where $\hat{D} = \sum_n (\boldsymbol{\epsilon} \cdot \mathbf{r}_n)$ is the operator of the interaction of electromagnetic radiation with the atomic system (in the dipole approximation) and summation is performed over all of the atomic electrons, $\boldsymbol{\epsilon}$ is the polarization vector of incident radiation, \mathbf{r}_n is the radius-vector of the photoelectron, s the intermediate state of the system with energy E_s , E_i is the atomic energy in the i state, and ω is the photon energy. In (1) and in what follows, atomic units are used ($\hbar = e = m_e = 1$), if not specified. Expression (1) is equivalent to

$$T_{i \rightarrow f}^{(2)} = \langle f | \hat{D} | \lambda \rangle + \langle \mu | \hat{D} | i \rangle - \langle \mu | E_i + \omega - \hat{H} | \lambda \rangle, \quad (2)$$

where

$$|\lambda\rangle = \frac{1}{E_i + \omega - \hat{H}} \hat{D} | i \rangle, \quad \langle \mu | = \langle f | \hat{D} \frac{1}{E_i + \omega - \hat{H}}, \quad (3)$$

and H is the atomic Hamiltonian. If we consider (2) as a functional that depends on the functions λ and μ , then it is stable relative to the variation of these functions. Therefore, the problem of obtaining amplitude (1) reduces to determining the unknown functions λ and μ that give the extremum of functional (2), while technically the method implies a simplification of (2) and developing the method of obtaining the functions λ and μ . The functionals of two functions for the amplitudes of high order are constructed in a similar way [9]. In this case, the wave functions of the initial state, i , and the final state, f , are assumed to be known.

Let us apply the variationally stable method to the process of two-photon ionization of a hydrogenlike system with charge Z . By separating the electron angular variables and integrating formula (2) in the matrix elements by these angles, we obtain the functional for the partial ionization amplitude into the channel with orbital momentum l_i of the electron

$$t_{i,l} = \langle \varepsilon_f l_f | r | \lambda_r \rangle + \langle \mu_r | r | n_i l_i \rangle - \langle \mu_r | E_i + \omega - \hat{h}(l) | \lambda_r \rangle. \quad (4)$$

Here, $\hat{h}(l) = -\frac{1}{2} \frac{d^2}{dr^2} - \frac{Z}{r} + \frac{l(l+1)}{2r^2}$ is the radial part of the Hamiltonian, $n_i l_i$ are the principal and orbital quantum numbers of the electron in the initial and final states, $\varepsilon_f l_f$ are the energy and orbital quantum

number in the continuous spectrum, l is the orbital momentum of electron in the intermediate state, and λ_r and μ_r are the radial parts of the unknown functions λ and μ . The integration in the matrix elements of formula (4) is performed over the radial coordinate. The reasoning does not change if instead of the Coulombic potential $-\frac{Z}{r}$ we substitute an arbitrary spherically symmetric potential $V(r)$. Functional (4) determines the absolute value of the amplitude in the case of real radial functions $R_{\varepsilon_f l_f}(r)$ for the state $\varepsilon_f l_f$ of the continuous spectrum. This is sufficient for calculating the integral values over the angles of the electron emission. For differential variables relative to the photoelectron emission angle, one must take the wave function with the phase in (4), which provides the asymptotics of an incoming spherical wave in the channel, l_f .

The problem of finding the extremum of functional (4) can be reduced to solving a system of equations, and with luck, a system of linear algebraic equations when choosing a suitable class of trial functions λ_r and μ_r on which the extremum of the functional is to be determined. We follow [5, 6] and others, in which the radial functions are expanded over the basis of Slater's orbitals $\Phi_m(r)$:

$$\lambda_r(r) = \sum_{m=1}^M a_m \Phi_m(r), \quad \mu_r(r) = \sum_{m=1}^M b_m \Phi_m(r), \quad (5)$$

where M is the number of Slater's orbitals, $\Phi_m(r) = N_m r^{\chi + m} e^{-\chi r}$, χ is still an arbitrary constant, N_m are the normalization multipliers. The main purpose of these multipliers is to avoid large numbers and computer overflow. The integral of the squared Slater's orbital is normalized to 1. The set of basis functions $\Phi_m(r)$ does not contain the exact wave functions of the hydrogen atom (ion) that could improve the convergence and future situations with many-electron atoms are modeled when the wave functions are unknown beforehand. For ease of analysis, the parameter χ is taken as common to all orbitals.

After substituting (5) in (4), the functional is transformed into the function with unknown coefficients a_m and b_m and obeys the conditions of extremality

$$\frac{\partial t_{l_f i}}{\partial a_m} = \frac{\partial t_{l_f i}}{\partial b_m} = 0. \quad (6)$$

By differentiating (4), one obtains the system of $2M$ linear algebraic equations for coefficients a_m and b_m

$$\sum_{m=1}^M A_{nm} a_m = c_n; \quad \sum_{n=1}^M b_n A_{nm} = d_m, \quad (7)$$

where $A_{nm} = \langle \Phi_n | E_i + \omega - \hat{h}(l) | \Phi_m \rangle$, $c_n = \langle \Phi_n | r | n; l_i \rangle$, and $d_m = \langle \varepsilon_f l_f | r | \Phi_j \rangle$. The use of Slater's orbitals $\Phi_m(r)$ with the constant parameter χ as the basis in

expansion (5) leads to a system of algebraic equations. Substituting coefficients a_m and b_m in (5) determines unknown functions λ_r and μ_r and, correspondingly, the partial amplitudes of two-photon transitions, (4). The procedure for obtaining functions λ_r and μ_r is repeated for each photon frequency and partial amplitude. Using the latter values, one obtains observable quantities in the process of two-photon ionization.

The main advantages of the variationally stable method include the absence of an infinite sum over intermediate states, the absence of an iteration procedure that leads to an increase in the errors for the amplitudes of higher order (as in the method by Dalgarno-Lewis [22]), the stability of the expression for amplitudes near the resonances, and the absence of zero energy differences in the denominator. In applications of the method, knowledge of the wave functions of initial and final states is sufficient, regardless of the amplitude order (the number of photons). In this case, the method is reduced to obtaining only two effective functions that are independent of the energy. Currently, only the lack of testing and as a consequence, the lack of information about the ranges of applicability are likely disadvantages of the variationally stable method.

2. CALCULATION RESULTS AND DISCUSSION

As a result of two-photon ionization of hydrogenlike atoms (ions) the electrons of the ground state $1s$ ($l_i = 0$) pass into the states of a continuous spectrum with orbital quantum numbers ($l_f = 0$) (according to the selection rules). In this case $l = 1$. As was shown in [9, 23], the amplitudes of multiphoton ionization of hydrogenlike ions obey scaling rules: they can be obtained from the amplitudes of a hydrogen atom by scaling the photon energy with the multiplier Z^2 and introducing the proportionality coefficient Z^4 before the amplitude. Therefore, the calculation of amplitudes for different Z values has meaning only in studying the numerical stability of the method. We carried out the calculations for the hydrogen atom ($Z = 1$) and the hydrogenlike ions of He ($Z = 2$) and neon ($Z = 10$) in the case of photon energies that are below the threshold of single-photon ionization. For linearly polarized radiation, the integral cross section of the two-photon ionization has the form [21, 23, 24]

$$\sigma = \frac{I}{I_0} \frac{2\pi^2 \alpha \omega}{45} (4|t_d|^2 + 5|t_s|^2), \quad (8)$$

where α is the fine-structure constant, I the intensity of incident radiation in W cm^{-2} , and $I_0 = 7.019 \times 10^{16} \text{ W cm}^{-2}$. We introduced the notations $t_s = t_{l_f=0, j=1}$, $t_d = t_{l_f=2, j=1}$. The amplitudes t_s and t_d were calculated using the variationally stable method with different values of parameters χ and M , as well as using the exact

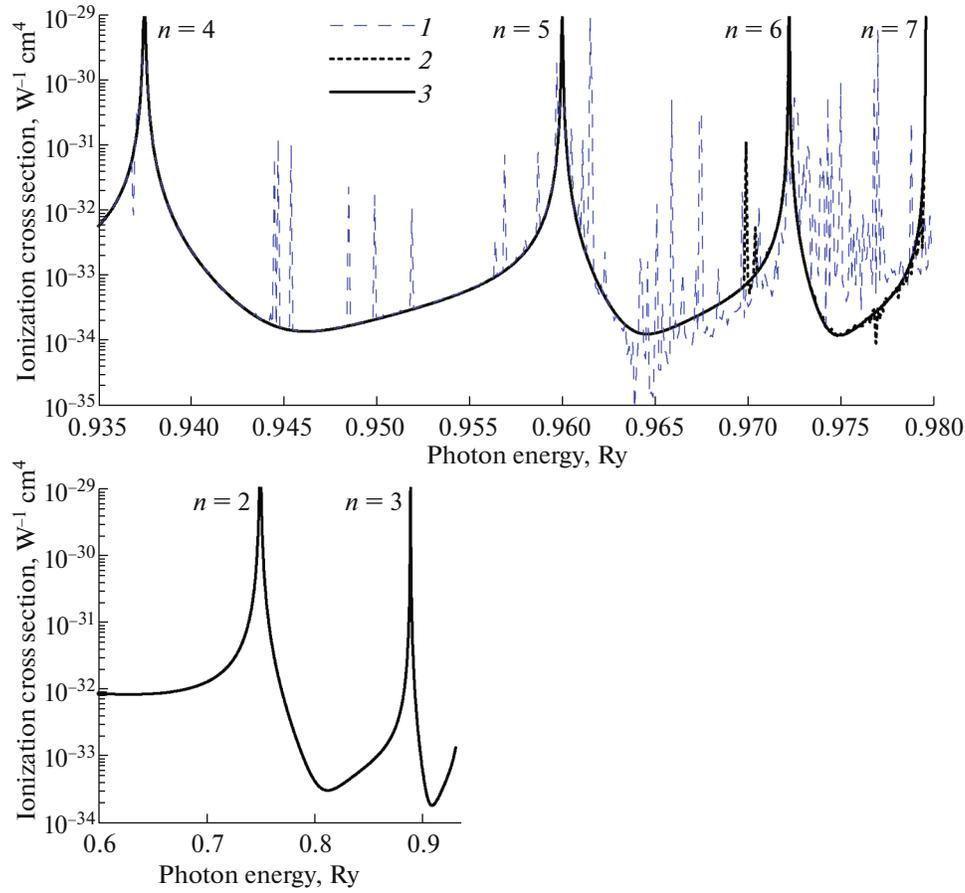


Fig. 1. The cross section of the two-photon ionization of the ground state of the hydrogen atom depending on the photon energy at $\chi = 0.5$ for two values of M : $M = 20$ (1) and $M = 40$ (2). Curve 3 shows an accurate result of the second-order perturbation theory. The three curves on the bottom panel are not distinguishable. The principal quantum numbers, np , of the resonances are indicated.

analytical formulas of the second-order perturbation theory [25]. Along with cross section (8), which has the dimensionality of length squared, but depends on the intensity of the field, the two-photon cross sections $\tilde{\sigma} = I^{-1}\sigma$ are also used, which depend only on the target characteristics with dimensionality $W^{-1}cm^4$.

First, we consider the convergence of the results with respect to the number M of Slater's orbitals in expansions (5). Figure 1 shows the calculated cross sections $\tilde{\sigma}$ of the two-photon ionization from the ground state of hydrogen atom ($E_i = -1$ Ry) at various M in (5) and fixed parameter $\chi = 0.5$. To estimate the accuracy of the method on the chosen class of trial functions (5), Fig. 1 shows an accurate cross section that is obtained via the second-order perturbation theory. The upper panel in Fig. 1 shows the magnified region of highly excited states of hydrogen atom. The cross sections contain sharp resonances that correspond to the positions of intermediate np states. At low photon energies, the total cross section is described by a small number of terms M in (5), which are in excel-

lent agreement with the exact result. Deviations from the exact results appear when approaching the photon energy to the ionization threshold. When the value of M is not large enough, the calculated cross section contains chaotic emissions, and with increasing M the region of emissions is shifted toward higher photon energies. This behavior of convergence can be explained by the fact that one needs to use an increasing number of Slater's orbitals to describe the oscillating wave functions of higher states np which, in addition, have different asymptotics at large distances from the nucleus. Although we presented the cross sections summed over the channels s and d , the regularities in the partial ionization cross sections turned out to be the same as we noted above in the case of the total cross section.

For a numerical illustration, the table shows the accurate cross sections of two-photon ionization of the ground state of the hydrogen atom that was obtained via the second-order perturbation theory and using the variationally stable method with $\chi = 0.5$ and $M = 40$. We chose the range of photon energies near

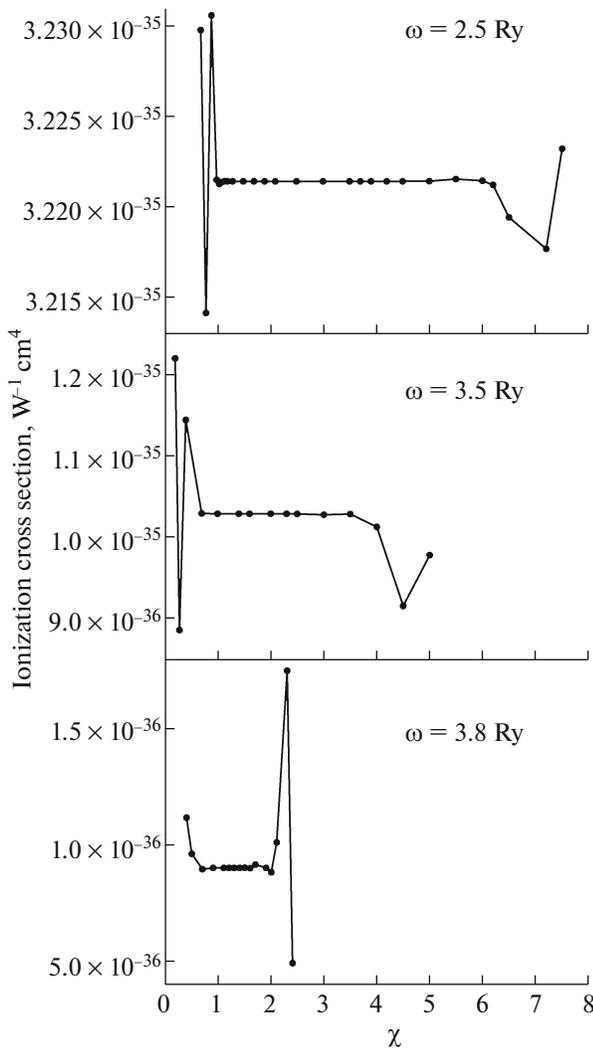


Fig. 2. The same as in Fig. 1 for the ground state of the helium ion He^+ at $M = 50$, obtained by the variationally stable method, depending on χ at three photon energies.

the resonance that correspond to the transitions through the intermediate $2p$ state of hydrogen atom ($\omega = 0.75$ Ry), where the magnitude of the cross section rapidly changes by several orders of magnitude.

Cross sections of two-photon ionization of a hydrogen atom (expressed in W^{-1}cm^4)

Photon energy, Ry	Exact value	Our calculation
0.770	9.19319×10^{-33}	9.19456×10^{-33}
0.760	5.53081×10^{-32}	5.53107×10^{-32}
0.751	7.68385×10^{-30}	7.68586×10^{-30}
0.749	8.22946×10^{-30}	8.23164×10^{-30}
0.740	1.10103×10^{-31}	1.10133×10^{-31}
0.730	3.69455×10^{-32}	3.69550×10^{-32}

For all energies, the results agree up to three or four significant digits.

We will now discuss the choice of the parameter χ based on the example of the calculation of the helium ion ionization cross section ($E_i = -4$ Ry), assuming the fixed parameter $M = 50$. Figure 2 shows the total cross sections of two-photon ionization of the ground state of the helium ion that depend on χ that corresponds to three fixed photon energies. In a certain range of the variation of the χ parameter, the cross sections remain nearly constant and correspond to the exact values. Out of this range, the results are not stable and differ from the accurate values. Therefore, the stability of the results relative to the variation of the parameter χ in the interval on the order of a unit can be considered as an indicator of incorrectly determined $t_{l,l'}$ amplitudes. In the range of high-order Rydberg states, the interval of the optimal χ values becomes narrower. Figure 3 shows the total ionization cross sections of the helium ion depending on the photon energies at $M = 50$ and various χ , and their comparison with accurate cross sections. The χ parameters were taken from the range of stability that were found above. The upper panel displays a magnified region of photon energies that are close to the energy threshold. For low photon energies, the cross sections of two-photon ionization for all χ values are nearly equal and agree with the exact calculations. For $\chi = 1.15$ the cross section agrees with the exact calculations in a wide range of photon energies, including the region of resonances up to $n = 6$. Even a small change in χ from 1.15 to 1.35 leads to considerable deterioration of convergence.

Using the variationally stable method, the analogous calculations were carried out for the Ne^{9+} ion. In this case, the best convergence was reached at approximately $\chi = 5.0$. Since in the Coulombic field the argument of wave functions is scaled with nucleus charge as Zr , the inverse proportionality of χ and the charge seems to be quite natural. Therefore, the empirical relationship $\chi \approx Z/2$ proves to be an optimal value for χ and with increasing Z the convergence of the method becomes slightly worse.

The angular distributions of photoelectrons, in contrast to the cross sections that are integrated over angles, depend on the interference between the partial amplitudes of ionization into s and d channels, i.e., on the relative phases of these amplitudes. Thus, for linear polarization of radiation, the angular distribution of photoelectrons is written in the form (for example, [26, 27])

$$\frac{d\sigma}{d\Omega} = \frac{\sigma}{4\pi} [1 + \beta_2 P_2(\cos\theta) + \beta_4 P_4(\cos\theta)] \quad (9)$$

$$= A + B \cos^2 \theta + C \cos^4 \theta,$$

where the angle θ is counted from the direction of polarization, $P_k(x)$ are the Legendre polynomials, A, B , and C are coefficients with the dimensionality of

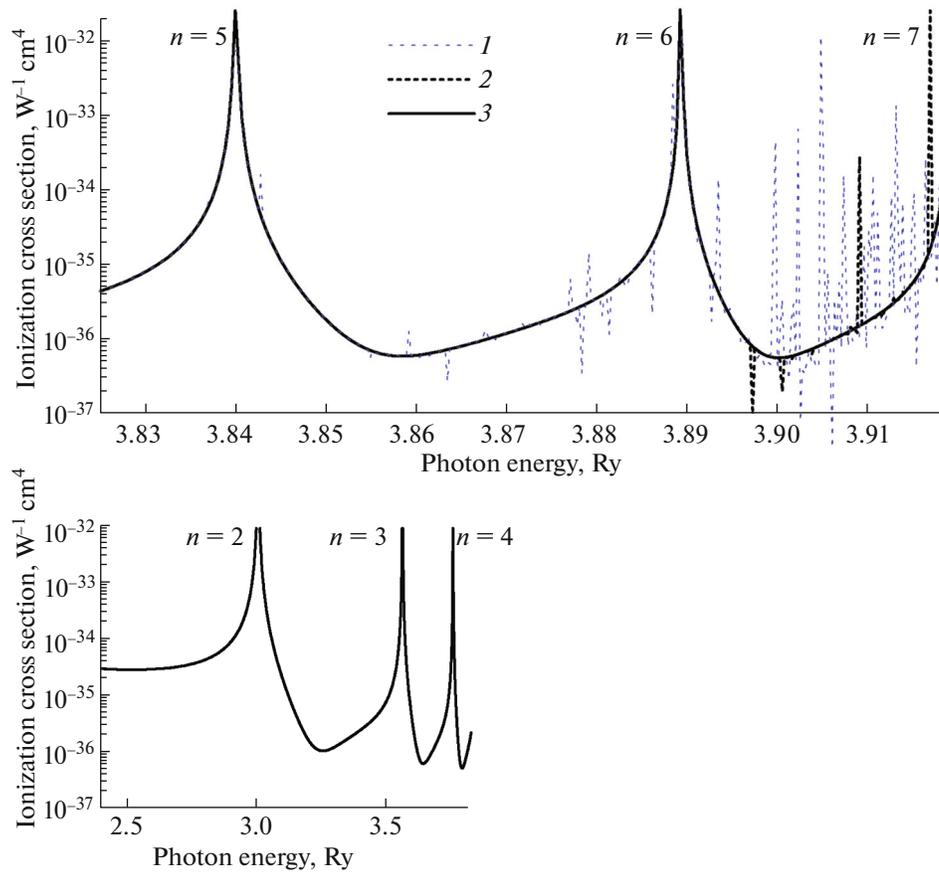


Fig. 3. The same as in Fig. 2 at $M = 50$ and two values of χ : $1 - \chi = 1.35$ (1) and $2 - \chi = 1.15$ (2). Curve 3 shows an accurate result of the second-order perturbation theory. The principal quantum numbers, np , of the resonances are indicated.

the cross section, β_2 and β_4 are the dimensionless coefficients of angular asymmetry. The latter coefficients are expressed via the partial amplitudes of ionization (4) [28]

$$\beta_2 = \frac{10}{W^2 + 1} \left[\frac{1}{7} - \frac{W}{\sqrt{5}} \cos \delta \right], \quad (10)$$

$$\beta_4 = \frac{18}{7(W^2 + 1)}. \quad (11)$$

Here $W = |t_s| \cdot |t_d|^{-1}$ is the ratio between the absolute values of the partial amplitude of two-photon ionization into the s and d channels, $\delta = \delta_s - \delta_d$ is the difference between the Coulombic phases, $\delta_l = \arg \Gamma \left(l + 1 + i \frac{z}{\sqrt{2\varepsilon_f}} \right)$.

Figure 4 shows the calculated parameters β_2 and β_4 of the angular anisotropy for two-photon ionization of the hydrogen atom. The parameters that were obtained in this work are in excellent agreement with the accurate values. The latter parameters (in the form of the coefficients A, B, C in (9)) were also given earlier in [26] for several values of the photon energy. Curves

1 and 2 for parameters β_2 and β_4 in Fig. 4 are universal: they are suitable for any hydrogenlike atom (in the nonrelativistic approximation) if the photon energies are taken in the units of $Z^2 E$. From (11) it always follows that $\beta_4 \geq 0$, but β_2 may take negative values due to the interference between the amplitudes of the ionization into the s and d channels. Both parameters of asymmetry are equal to zero if the ionization into the d channel is absent. This is natural, since the s wave alone may give only an isotropic angular distribution. In the case where the ionization into the s channel is completely suppressed, the β_4 parameter takes its maximum value $\beta_4 = 18/7$, and β_2 takes a fixed value of $10/7$. The behavior of the curves that show the asymmetry of the parameters clearly indicates the photon energies at which a particular ionization channel dominates.

Convergence of the calculations of angular distributions with increasing M in the range of highly excited states is demonstrated on the upper panel in Fig. 4 based on the example of the β_2 parameter of asymmetry. Comparison of Fig. 4 and Fig. 1 clearly shows that the most of random emissions in the cross sections are absent in the β_2 parameter. These emis-

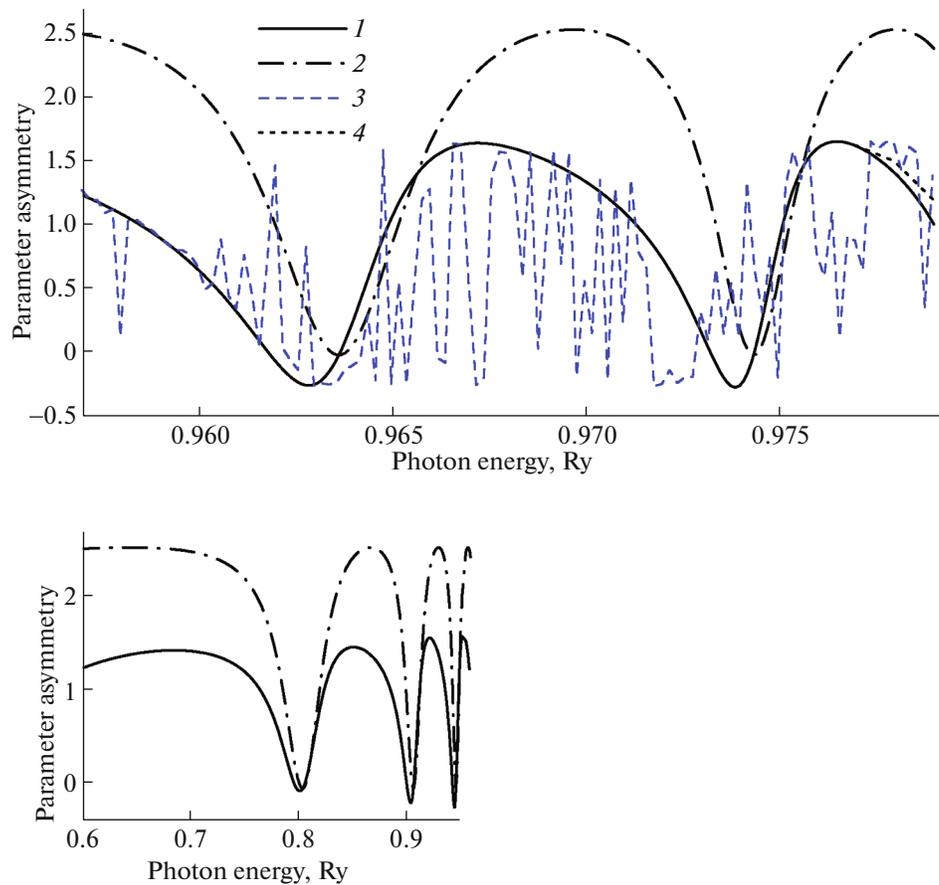


Fig. 4. The β_2 and β_4 parameters of the angular anisotropy for two-photon ionization of hydrogen atom depending on the photon energy. Curves 1 and 2 correspond to β_2 and β_4 that were obtained using the second-order perturbation theory. Curves 3 ($M = 20$) and 4 ($M = 40$) correspond to β_2 that was obtained using the variationally stable method at $\chi = 0.5$. Curves 1, 3 and 4 are not distinguishable on the bottom panel.

sions are mutually cancelled in the amplitudes of ionization in the s and d channels, which leads to a much better convergence of the anisotropy parameter than the cross section.

CONCLUSIONS

In this work, we presented the basic formulas for calculating the characteristics of the two-photon ionization in the framework of the variationally stable method and analyzed the choice of the parameters of trial functions. The calculation results of the photoionization cross sections and parameters of angular anisotropy for hydrogen atom and hydrogenlike ions are given. Comparison of the obtained and accurate results shows that by varying the parameters of the trial functions one can calculate the characteristics of the two-photon ionization of the hydrogen atom and hydrogenlike ions with a good accuracy for a wide range of photon energies, including the ranges of resonances. After proper generalization, we plan to use the results and the software for calculating the characteristics of two-frequency processes and the above-

threshold ionization of multielectron atoms in the range of VUV and soft X-rays.

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