The influence of the resonance effects on the radiative characteristics of helium plasma

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Abstract. In the present work, the influence of an alternating circularly polarized electric field on the energy spectrum of the He atom is studied. The calculations are performed by the method of the energy matrix diagonalization of an atom in the electric field. This method has allowed us to study the behaviour of the helium energy spectrum from the same numerical procedure under resonant and non-resonant excitations by the electric field. Based on the calculation results, we have found that the resonance effects take place not only in the vicinity of resonance, but they influence the shift directions of the Stark states even under non-resonant excitation. Additionally, we have established that the helium energy spectrum behaves consistently in the electric field. The results obtained have allowed us to clarify mechanisms of the influence of the resonance effects on the radiative characteristics of helium plasma.

1. Introduction
The study of the influence of an alternating electric field on atomic spectra is a topical problem of modern physics. After first experiments, where the dynamic Stark effect was detected, general theoretical methods were developed for calculating the interaction of laser fields with atoms [1, 2 and the references therein]. In the framework of these methods based on perturbation theory, formulas for calculating the Stark state shifts in electric fields of different polarization were derived [1, 3 and the references therein]. All formulas in these papers were derived for the case of non-resonant excitation by electric fields. In the case of resonant excitation, it is necessary to use the resonance approximation [4, 5]. Both for resonant and non-resonant excitations, limitations of non-stationary perturbation theory (perturbation by the electric field is not very great, atomic levels are isolated, and atoms under consideration are Rydberg ones) must be satisfied [1 – 3, 5]. In so doing, by virtue of computational complexity in the framework of perturbation theory, one has to restrict oneself to the two-, three-, and maximum four-level approximations in calculating the AC Stark effect [6, 7].

By the present time, a lot of other excitation sources (in particular, high-frequency electrodeless discharge lamps, light-emitting diodes, superpower lasers, and so on) have appeared. These sources generate electric fields with parameters essentially different from those of optical lasers. Atomic energy spectra induced by these electric fields do not satisfy above-mentioned conditions of validity of perturbation theory. Thus, a new theoretical approach is necessary for calculating atomic spectra excited by alternating electric fields.
In the present work, a theoretical method free from limitations of perturbation theory is used for calculating the helium emission spectrum in an alternating circularly polarized electric field. This numerical method suggested and developed by us in [8, 9] is based on diagonalization of the energy matrix of an atom in the electric field. The suggested approach is valid both under resonant and non-resonant excitations by the electric field, and allows us to take into account the Stark state interactions.

2. Calculation method

In a circularly polarized electric field, the non-stationary Schrödinger equation is written as

\[ i \frac{\partial \psi_n(r,t)}{\partial t} = (H_0(r) - F(x \cos \omega t \pm y \sin \omega t))\psi_n(r,t), \tag{1} \]

where \( \psi_n \) is the wave function of the \( n \)-th state of the system, \( H_0 \) is the unperturbed Hamiltonian, and the operator \(- F(x \cos \omega t \pm y \sin \omega t)\) describes perturbation induced by the interaction of an atom with a circularly polarized electric field of frequency \( \omega \) and strength \( F \). The “+” and “−” signs correspond to the right and left polarization of the field, respectively. For the case of a circularly polarized field, the non-stationary Schrödinger equation can be reduced to the stationary one due to separation of spatial and time variables within the framework of the rotating-wave approximation [10].

Within this approximation, the wave function in the coordinate system rotating about the Z-axis with the frequency \( \omega \), has the form

\[ \varphi(r,t) = \exp(i\omega J_z)\psi(r,t), \tag{2} \]

where \( J_z \) is the z-component of the total angular momentum operator. Substituting the equation (2) in the equation (1), we get

\[ i \frac{\partial \varphi(r,t)}{\partial t} = Q\varphi(r,t), \quad Q = (H_0 - \omega J_z \pm FD_z). \tag{3} \]

As seen from the equation (3), the operator \( Q \) is time-independent. Hence, in the rotating-wave approximation, it is possible to go from the non-stationary Schrödinger equation (1) to the stationary one, and we have

\[ Q\varphi(r) = \epsilon \varphi(r), \quad \varphi(r,t) = \exp(-i\epsilon t)\varphi(r), \tag{4} \]

where \( Q \) is the energy operator of an atom in the electric field, and \( \epsilon \) and \( \varphi(r,t) \) are the energy and wave function of an atom in the electric field in the rotating coordinate system. It is obvious that the Schrödinger equation (4) can be solved within stationary perturbation theory, but this way of solution has been found too complicated [1, 2, 11]. We have found that instead of solving the Schrödinger equation (4) within a perturbation theory, it is much more convenient to solve it using a diagonalization procedure. It was shown that the wave functions and energies of an atom, being solutions to the Schrödinger equation (4), can be found from diagonalization of the energy matrix of an atom in the electric field with the following elements

\[ Q_{nn'} = E^{(n)}_n \delta_{nn'} - \omega < \phi^{(n)}_m(r) | J_z | \phi^{(n)}_n(r) > \pm F < \phi^{(n)}_m(r) | D_x | \phi^{(n)}_n(r) >. \tag{5} \]

In equation (5), \( \phi^{(n)}_m \) and \( E^{(n)}_n \) are the wave function and energy of the \( n \)-th state of an atom in the absence of the electric field; \( J_z \) is the z-component of the total angular momentum operator; and \( D_x \) is the x-component of the dipole moment operator. The matrix elements of the \( D_x \) operator are determined as

\[ < \phi^{(n)}_m | D_x | \phi^{(n)}_{n'} > = \chi[M,D,J,J'] = \frac{(-1)^{J-M}}{2} \left[ \begin{array}{cc} J & 1 \\ -M & 1 \end{array} \right] \left[ \begin{array}{cc} 1 & J' \\ 1 & -M \end{array} \right] \chi[D,J,J'], \tag{6} \]

Details of calculating the matrix elements (6) are reported in [9, 12, 13].
The energy matrix with the elements (5) is obtained in the coordinate system rotating about the Z-axis with frequency $\omega$. Upon diagonalization of this matrix, we get the energies $\varepsilon_n$ and wave functions $\varphi_n$ as

$$\varphi_n(r, t) = \exp(-i\varepsilon_n t) \sum C_{nk}(F, \omega)\varphi_k^{(0)}(r)$$

for the $n$ atomic states in the electric field in the rotating coordinate system. The coefficients $C_{nk}$ in the wave function (7) are the expansion coefficients of the wave functions of an atom under the action of the electric field in terms of unperturbed wave functions. These coefficients depend on the electric field strength and frequency.

To go to the initial coordinate system and find the average atomic energies in the electric field, it is necessary to perform averaging over the oscillation period of the electric field. After this averaging, the wave functions and average energies of an atom in the electric field in the initial coordinate system are written as

$$\psi_n(r, t) = \exp(-i(\omega J_x + \varepsilon_n)t) \sum C_{nk}(F, \omega)\varphi_k^{(0)}(r),$$

$$E_n = \langle \psi_n(r, t)|H(r, t)|\psi_n(r, t) \rangle \approx \varepsilon_n + \omega < \varphi_n(r)|J_x|\varphi_n(r).$$

It follows from the above reasoning that the proposed theoretical approach is free from limitations of perturbation theory and can be used for calculating the dynamic Stark effect in a circularly polarized electric field with the strength and frequency changing in wide ranges. Since the energies and wave functions in the electric field are determined by the diagonalization procedure, it allows us to take into account the Stark state interactions. The algorithm of our theoretical approach is implemented in a special software package StarkD [14].

In this work, the described above method is applied to the study of the dependence of the helium energy spectrum on the electric field frequency.

3. Results and discussion

It is well-known from the perturbation theory that the Stark shifts of atomic states induced by an alternating circularly polarized electric field are calculated as

$$\Delta E = -\frac{1}{2} \alpha F^2,$$

where $\alpha$ is the the polarizability of an atom, and $F$ is the electric field strength. If the electric field frequency equals the transition frequency between the Stark states, the curve characterizing the dependence of the polarizability on the electric field strength has resonances. In these resonances and near-resonance ranges, the polarizability calculated using the formulas obtained in [1 – 3] is infinite, therefore, in the case of resonant excitation by the electric field, the polarizability $\alpha$ must be calculated in the resonance approximation [4, 5, 15]. After the transition through resonance, the polarizability changes in the sign [16]. Hence, the Stark state shifts have big values in resonances, and, by virtue of equation (10), the Stark state shifts change their directions after the transition through resonances.

Since the influence of the electric field leads to mixing the Stark states, it is desirable to take into account the Stark state interactions in calculating atomic spectra induced by electric fields. In the framework of our numerical method, taking into account of these interactions occurs automatically, because the wave functions and energies of atoms in the electric field are determined by the diagonalization procedure.

Let us consider the results of calculating the $\omega$-dependence of the shift directions of the Stark states obtained in the framework of our theoretical method for the He atom.

First of all, we have carried out the calculations of the helium spectrum under resonant excitation by the electric field. In the framework of our theoretical approach, there are no difficulties in calculating the Stark state shifts just in resonances. The calculation results for the $S$ state shifts of the He atom in resonances are listed in table 1. In this table, the Stark state shift is defined as $\Delta E = E - E_0$. 

where $E$ is the energy of an atom in the electric field calculated using equation (9), and $E_0$ is the energy of this atom in the absence of the electric field. The values of the electric field frequency in the second column are the resonance frequencies of the corresponding transition from the first column. The Stark state shifts in resonances have been computed at the electric field strength $F=1.2$ kV/cm. It should be noted that the $6^3S_1$ energy level as well as the $1^1S_0$ levels does not split in the electric field, because this level weakly interacts with neighbouring energy levels when the electric field strength is so small.

### Table 1. Shifts of the $S$ states of the He atom in resonances ($F=1.2$ kV/cm).

| Resonance | $\omega$ (MHz) | $\Delta E$ (cm$^{-1}$) |
|-----------|----------------|-------------------------|
| $1s^2\,^1S_0 - 2\,^1P_1$ | 5.1035·10$^9$ | 5906.058 |
| $1s^2\,^1S_0 - 3\,^1P_1$ | 5.5820·10$^9$ | 27456.021 |
| $1s^2\,^1S_0 - 4\,^1P_1$ | 5.7408·10$^9$ | 11022.708 |
| $6\,^1S_0 - 6\,^1P_1$ | 0.47979·10$^7$ | 7.85978 |
| $6\,^1S_0 - 7\,^1P_1$ | 2.89066·10$^7$ | 480.4672 |
| $6\,^3S_1 - 6\,^3P_2$ | 7.69337·10$^6$ | 126.8043 |
| $6\,^3S_1 - 6\,^3P_0$ | 7.69345·10$^6$ | 126.9727 |
| $6\,^3S_1 - 6\,^3P_0$ | 7.69440·10$^6$ | 127.6617 |

As it follows from table 1, the Stark state shifts in resonances have finite values, and there is no necessity to use the resonance approximation.

In order to compare the $\omega$-dependence of the Stark state shifts obtained by our numerical method with those calculated in the framework of non-stationary perturbation theory, we have plotted the curves characterizing dependences of the ground and excited $S$ states of the He atom on the electric field frequency. These curves calculated at the electric field strength $F=1.2$ kV/cm are shown in figures 1–3. As it is seen from these figures, when the electric field frequency equals the resonance frequency, we get a drastic increase in the Stark state shifts in resonances and near-resonance ranges.

This result agrees with the perturbation theory that confirms a validity of our theoretical approach. It is interesting to note that peaks characterizing the Stark state shifts in resonances are very sharp for singlet states (see figures 1 and 2, and table 1), whereas these peaks are not very high for triplet states of the He atom (see figure 3).

Now, let us consider the case of non-resonant excitation of the helium spectrum. The calculations of the helium state shifts are carried out for different frequencies of the electric fields generated by real excitation sources, namely, $\omega=100$ MHz (a high-frequency discharge HFD), $\omega=1.5191\cdot10^5$ MHz (a
NH₃-laser), $\omega=2.4352\cdot10^6$ MHz (a HCN-laser) and $\omega=2.4181\cdot10^7$ MHz (a CO₂-laser). The calculation results for singlet and triplet S states of the He atom are shown in figures 4 and 5. These figures demonstrate that an increase in the electric field frequency leads to the resonance effects (that is, to the changes of the shift directions of the Stark states) even under non-resonant excitation by the electric field. Based on the calculation results, we can conclude that the presence of the resonance effects results from two reasons.

![Figure 4](image1.png)

**Figure 4.** Dependence of the Stark state shifts on the electric field frequency for the $1sns^1S_0$ energy levels.

![Figure 5](image2.png)

**Figure 5.** Dependence of the Stark state shifts on the electric field frequency for the $1sns^3S_1$ energy levels.

Firstly, an increase in the electric field strength leads to restructuring of the energy spectrum, that is, to changing the positions of the Stark states relative to each other. To illustrate restructuring of the helium spectrum in the electric field, we list energy state numbers for the $^1S_0$ and $^3P_0$ states of the He atom for different $\omega$ (see table 2). It follows from this table that owing to restructuring of the energy spectrum the transition frequencies change with an increase in the electric field frequency and it leads to appearance of resonances.

| Level | $\omega_{\text{HFD}}$ | $\omega_{\text{NH}_3}$ | $\omega_{\text{HCN}}$ | $\omega_{\text{CO}_2}$ |
|-------|-----------------|-----------------|-----------------|-----------------|
| $^1S_0$ | 5s   | 121  | 121  | 123  | 158  |
|       | 6s   | 185  | 185  | 200  | 232  |
|       | 7s   | 249  | 249  | 264  | 294  |
|       | 8s   | 313  | 313  | 336  | 347  |
| $^3P_0$ | 5p   | 130  | 127  | 139  | 162  |
|       | 6p   | 192  | 191  | 211  | 235  |
|       | 7p   | 258  | 255  | 276  | 299  |
|       | 8p   | 322  | 319  | 332  | 350  |
Secondly, since the wave functions and energies of atoms in the electric field are determined by the diagonalization procedure, all atomic states included in the energy matrix behaves consistently, that is, if the action of the electric field leads to changing the shift directions of the np-states, then the n’s- and n’d-states also change their shift directions. The reason for such consistent behavior is the fact that, according to equation (5), all Stark states in the helium spectrum are linked by the matrix elements of the x-component of the dipole moment operator $D_x$.

As an illustration of a consistent behavior of the helium spectrum in the electric field, in addition to figures 4 and 5, we present figures 6 and 7. These figures show the dependence of the np state shifts on the electric field frequency for the He atom. In figure 7, the values of the magnetic quantum number $M$ for the $7^1P_1$ level are presented in a black frame. Comparison of figures 5 and 6 illustrates a consistent change in the shift directions of the $8^3S_1$ and $7^3P_0$ levels of the He atom.

As it is seen from figures 4 – 7, for the np states as well as for the ns states, non-resonant excitation by the electric field leads to drastic changes in the shift directions of the Stark states. This phenomenon is obtained for the energy levels both non-splitting (see figures 4 – 6) and splitting (see figure 7) in the electric field. For the energy levels, non-splitting in the electric field, the resonance effects result in the breakdown of a monotonous dependence of the Stark state shifts on the principal quantum number $n$ of the outer-shell electron (see figures 4 – 6). For the energy levels, splitting in the electric field, the presence of the resonance effects also leads to changing the shift directions of the Stark states (see figure 7).

Thus, the calculations carried out in the framework of our theoretical method allow us to reveal the reasons for the changes in the shift directions of the Stark states of the He atom under non-resonant excitation by the electric field. It is obvious that these changes influence the radiative characteristics of helium plasma, in particular, they lead to changing the colour characteristics of plasma emission. Changing the colour characteristics of lamps with an increase in the electric field frequency was observed in experiments [17 and references therein].
4. Conclusions
In this paper, it is shown that the method of the energy matrix diagonalization of an atom in the electric field enables to carry out calculating atomic emission spectra by the diagonalization procedure both under resonant and non-resonant excitations by the electric field. In addition, it has been found that the resonance effects take place under non-resonant excitation of the He atom, and restructuring of the energy spectrum is the reason for these effects. In so doing, the emission spectrum of the He atom behaves consistently. Finally, it has been shown that the resonance effects are the reasons for changing the shift directions of the Stark states under non-resonant excitation by the electric field.

These theoretical results obtained for the first time allow us to clarify mechanisms of the formation of the helium spectrum in the electric field and have practical applications in a plasma spectroscopy, a gas discharge physics and a laser physics.

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