Software package StarkD for calculating atomic emission spectra in an alternating electric field

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Abstract. In the present work, the software package StarkD for calculating the emission spectra of atoms in an alternating circularly polarized electric field is presented. The emission spectra are calculated by the numerical method based on diagonalization of the energy matrix of an atom in an electric field. This method is free from limitations of perturbation theory and valid in wide ranges of the electric field frequency and strength. The package StarkD allows us to calculate the positions of the Stark components of atomic spectral lines as well as the transition probabilities, line intensities, and spectral line profiles of atoms in the electric field. The package capabilities are illustrated by the examples of calculating the emission spectra of He, Ne, Ar, and Kr atoms in the electric field. The calculations have allowed us to establish a number of interesting theoretical regularities and to analyze mechanisms of the formation of atomic spectra in the electric field.

1. Introduction

A theoretical study of the influence of an alternating electric field on atomic spectra is a topical problem of modern physics. Calculating the spectroscopic characteristics of atoms in the electric field is interest of many branches of physics, namely: gas discharge physics, magnetic reconnection, laser physics, plasma diagnostics, and so on. General theoretical methods for calculating the interaction of laser fields with atoms were developed at the end of previous century [1, 2 and the references therein]. In the framework of these methods based on non-stationary perturbation theory, formulas for calculating the Stark state shifts, polarizabilities and hyperpolarizabilities for atoms in electric fields of different polarization were derived [1, 3 and the references therein]. Non-stationary perturbation theory is valid only in the case, when the following limitations are satisfied: perturbation by the electric field is not very great, atomic levels are isolated, and atoms under consideration are Rydberg ones [1 – 4]. Moreover, 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 [4 – 6].

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. By virtue of its limitations, non-stationary perturbation theory is not suitable for the above-listed excitation sources.
In addition, in astrophysics [7], in solving problems of plasma diagnostics [8], in the investigation of magnetic reconnection [9], and in technologies of creation of new radiation sources [10], an urgent need arose in the knowledge of various regularities in the behavior of the spectroscopic characteristics of atoms in electric fields with the parameters changing in wide ranges. Thus, for solving the above-mentioned problems, one needs a new theoretical approach for calculating atomic spectra excited by alternating electric fields.

In the present work, a theoretical method free from limitations of perturbation theory is proposed for calculating atomic emission spectra in an alternating circularly polarized electric field. This numerical method developed by us in [11] is based on diagonalization of the energy matrix of an atom in the electric field. The suggested approach allows us to take into account the interaction of all Stark states included in the energy matrix, and it is valid for calculating atomic emission spectra in a circularly polarized electric field with the electric field strength and frequency changing in wide ranges. The algorithm of the developed theoretical method is implemented in a special software package StarkD. The rare gas atoms are chosen as the object of study, because these gases are very often used in different lamps, light sources and other devices, and a theoretical calculation of the spectroscopic characteristics of these atoms in the electric field is a vital problem of modern physics.

2. Calculation method

In the case of an alternating circularly polarized electric field, the energies and the wave functions of an atom in the electric field are determined from the non-stationary Schrödinger equation

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

where \( H_0(r) \) is the Hamiltonian of the unperturbed system, and the operator \( F(x \cos \omega t \pm y \sin \omega t) \) describes perturbation of an atom by the circularly polarized electric field of frequency \( \omega \) and strength \( F \). In the framework of our method of the energy matrix diagonalization of an atom in an electric field, we solve Eq. (1) using the two-step procedure.

In the first step, the non-stationary Schrödinger equation given by Eq. (1) is transformed into the stationary one. Such transformation can be realized in the rotating-wave approximation [12]. In this approximation, the wave function of an atom in the coordinate system rotating about the Z-axis with the electric field frequency \( \omega \) is written as

\[ \psi(r,t) = \phi(r,t) \exp(i \omega t J_z), \]

where \( J_z \) is the z-component of the total angular momentum operator. After substitution of the wave function given by Eq. (2) in Eq. (1), we get the stationary Schrödinger equation in the form

\[ i \frac{\partial \phi_n(r,t)}{\partial t} = \varepsilon_n \phi_n(r,t), \]

\[ \phi_n(r,t) = \exp(-i \varepsilon_n t) \phi_n(r). \]

In Eqs. (3) and (4), \( Q \) is time-independent operator of the energy of an atom in the electric field; \( \varepsilon_n \) and \( \phi_n(r,t) \) are the energy and the wave function of the \( n \)th state of an atom in the electric field in the rotating coordinate system.

It should be noted that numerous attempts to solve Eq. (3) in the framework of stationary perturbation theory turned out to be unsuccessful by virtue of limitations of this theory and computational difficulties [1, 2]. Therefore, instead of using stationary perturbation theory, we solve Eq. (3) by diagonalization of the energy matrix \( Q \) with the matrix elements calculated in the representation of unperturbed wave functions \( \phi_n^{(0)} \). In this case, the atomic energies \( \varepsilon_n \) and the wave functions \( \phi_n(r) \), being solutions of the Schrödinger equation (Eq. (3)), are determined by diagonalization of the energy matrix \( Q \) with elements

\[ Q_{mn} = E_n^{(0)} \delta_{mn} - \omega < \phi_m^{(0)}(r)|J_z|\phi_n^{(0)}(r)> \pm F < \phi_m^{(0)}(r)|D_z|\phi_n^{(0)}(r)>, \]

\[ \phi_n^{(0)}(r) = \phi_n(r,t) \exp(i \omega t J_z), \]

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where \( \psi_n^{(0)} \) and \( E_n^{(0)} \) are the wave function and the energy of the \( n \)th state of an atom in the absence of the electric field and \( D_x \) is the \( x \)-component of the dipole moment operator. The matrix elements of this operator are

\[
\langle \psi_n^{(0)} | D_x | \psi_n^{(0)} \rangle = \gamma J M | \gamma J' M' \rangle \approx \left( \frac{-1}{\sqrt{2}} \right) \left[ \begin{pmatrix} J & 1 & J' \\ -M - 1 & M' \end{pmatrix} - \begin{pmatrix} J & 1 & J' \\ -M & M' \end{pmatrix} \right] \langle \gamma J | D_x | \gamma J' \rangle.
\]

The reduced matrix elements \( \langle \gamma J | D_x | \gamma J' \rangle \) in Eq. (6) depend on the type of coupling scheme. The energy states of rare gas atoms are well described in the \( LS \) coupling scheme (the He atom) and in the \( JL \) coupling scheme (Ne, Ar, and Kr atoms). Calculation formulas for \( \langle \gamma J | D_x | \gamma J' \rangle \) are

\[
\langle \gamma J | D_x | \gamma J' \rangle = Q(T_{ij}, T_{i'j'}) < R_{nl} | \gamma J' > ,
\]

\[
< R_{nl} | \gamma J' > = \int_0^\infty R_{nl}(r) r^2 dr ,
\]

where \( T_{ij}, T_{i'j'} \) are the quantum numbers of atomic energy levels in the above-mentioned coupling schemes. Calculation formulas for the multipliers \( Q(T_{ij}, T_{i'j'}) \) from Eq. (7) are reported in [13]. The main problem in calculating the matrix elements \( \langle \psi_n^{(0)} | D_x | \psi_n^{(0)} \rangle \) is the calculation of radial integrals given by Eq. (8). These integrals can be computed by different methods. A direct calculation of radial integrals with the analytical functions \( R_{nl} \) determined by any variational methods takes too much time. The calculation of the integrals \( < R_{nl} | \gamma J' > \) with the numerical wave functions determined by solving the Hartree–Fock equations is also extraordinarily cumbersome and takes a plenty of time. The well-known semiempirical method by Bates and Damgaard [14] allows us to compute the radial integrals more quickly, but the results of this method are not always correct.

In the framework of our approach, we propose an efficient and fast technique to calculate the radial integrals, where disadvantages of the method by Bates and Damgaard have been corrected. The specific form of our semiempirical formula for calculating the radial integrals and details of its derivation were reported in [11]. In order to verify the correctness of our method of calculating the radial integrals given by Eq. (8), we compared the radial integrals calculated by our fast method with those integrals computed with the analytical functions and the Hartree–Fock functions. The comparison has indicated good agreement of the radial integrals computed by all three methods that confirms the reliability of our technique.

In the second step of our procedure, after averaging over the oscillation period of the electric field, we get the wave functions and the average energies of an atom in the electric field in the initial coordinate system in the form

\[
\psi_n(r,t) = \exp(-i(\omega J_z + \varepsilon_n)t) \sum_k C_{nk} \psi_k^{(0)}(r) ,
\]

\[
\overline{E}_n = \langle \psi_n(r,t) | \mathbf{H}(t) | \psi_n(r,t) \rangle = \varepsilon_n + \omega \langle \psi_n(r) | J_z | \psi_n(r) \rangle ,
\]

where \( C_{nk} \) are the expansion coefficients of the wave functions of the examined atom under the action of the electric field in terms of the unperturbed wave functions \( \psi_k^{(0)}(r) \).

The wave functions given by Eq. (9) and the energies given by Eq. (10) are further used for calculating any atomic characteristics being of interest for spectroscopic analysis. In particular, in solving problems of plasma diagnostics, gas discharge physics, and astrophysics, one needs to calculate spontaneous transition probabilities between the Stark states and spectral line intensities in the emission spectra excited by the electric field. The expression for calculating the spontaneous transition probabilities between the Stark states \( J M \) and \( J'M' \) has the form
\[
A(JM \rightarrow J'M') = \frac{4\omega_{JM,J'M'}^3}{3hc^3} \sum_q \sum g C_j^{(JM)} C_j^{(J'M')} (-1)^{J_i-M_i} \left( \begin{array}{ccc} J_i & 1 & J_j \\ -M_i & q & M_j \end{array} \right) <\gamma J_i \| D \| \gamma J_j >^2 ,
\]
(11)

where \(C_j^{(JM)}\) and \(C_j^{(J'M')}\) are the expansion coefficients from Eq. (9) and \(\omega_{JM,J'M'}\) is the frequency of the \(JM\rightarrow J'M'\) transition. The reduced matrix elements \(<\gamma J \| D \| J' >\) in Eq. (11) are calculated using formulas (7) and (8).

The intensities of the Stark components of spectral lines are calculated as

\[
I(JM \rightarrow J'M') = N_{JM} A(JM \rightarrow J'M')(\overline{E}_{JM} - \overline{E}_{J'M'}) ,
\]
(12)

where \(N_{JM}\) is the population of the Stark state. The problem of calculating the Stark state population \(N_{JM}\) is very complicated, because exact values of \(N_{JM}\) are unknown, and these quantities can be evaluated only in the framework of some approximations. A choice of a concrete approximation is specified by excitation mechanism of a discharge. For thermodynamic equilibrium and LTE plasmas, the Stark state populations are estimated from the Boltzmann distribution. In case of the non-equilibrium emission spectrum, one can use the static and dynamic approximations for estimating the Stark state populations [15, 16]. In the framework of our theoretical approach, the Stark state populations can be computed using any one of these approximations.

Based on the calculated intensities of the Stark components, it is possible to obtain the total profile of a spectral line specified by the influence of the electric field and other mechanisms of broadening of spectral lines. The calculation of profiles of the Stark components of atomic spectral lines is performed using the formula

\[
I(\nu) = I_0 \phi(\nu) ,
\]
(13)

where \(I_0\) is the intensity of the Stark component in its maximum calculated using Eq. (12), and \(\phi(\nu)\) is the line profile function specifying the type of the line profile, namely: the Lorentz, the Doppler or the Voigt profiles. In the framework of our theoretical approach, all these profiles can be calculated. The total profile of a spectral line is the sum of the calculated profiles of the Stark components of this spectral line.

So, it follows from the above reasoning that no one formula of the proposed theoretical approach has limitations on the parameters of the electric field inherent in perturbation theory. Moreover, since we determine the energies and the wave functions of atoms in the electric field by diagonalization procedure, in the framework of our approach the interaction of all Stark states is taken into account automatically, whereas in perturbation theory, atomic states are considered as isolated ones. Therefore, the suggested numerical approach for calculating atomic spectra in the electric field is free from limitations of perturbation theory and can be used for calculating atomic emission spectra in a circularly polarized electric field with the electric field strength and frequency changing in wide ranges.

3. Software package StarkD

The theoretical approach described in Section 2 has been implemented in a special software package StarkD written in Fortran and Maple. A flow diagram of this package is given in Fig. 1. As seen from this diagram, the package StarkD is divided into two parts. The first part of the package gets us the wave functions and the average energies of an atom in the electric field. The second part of the package calculates different spectroscopic characteristics of atoms in the electric field using the wave functions and the energies obtained in the first part. Let us consider both parts of the software package StarkD.

3.1. Part 1 of the package StarkD. Programs for calculation and diagonalization of the energy matrix of an atom in the electric field.

All programs of this part of the package StarkD are written in Fortran.
DATA INPUT: inputs the electric field parameters $F$ and $\omega$, and the positions of unperturbed energy levels with their quantum numbers for the examined atom. To input these data, a graphical user interface is used.

RAD: computes the radial integrals given by Eq. (8). The calculations of these integrals are performed using our efficient and fast technique (see discussion in Section 2).

DIPOL: computes the matrix elements of the $D_x$ operator given by Eq. (6). To calculate the reduced matrix elements $<J'\mid D_x \mid J>$, additional programs for calculating the Clebsch-Gordan coefficients and the $nj$-symbols ($n=3, 6, 9, 12$) are written.

QNM: computes the matrix elements of the energy matrix given by Eq. (5).

DIAGL: diagonalizes the energy matrix of an atom in the electric field. Diagonalization procedure is thoroughly tested. The performance index $\tau$ is used as the criterion of the procedure accuracy. If the matrix equation

$$Ax = \lambda x,$$  \hspace{1cm} (14)

is solved, then the performance index is defined to be

$$\tau = \max_{1\leq j \leq n} \frac{\| A\tilde{x}_j - \tilde{\lambda}_j \tilde{x}_j \|}{10^{-n} \| A \| \| \tilde{x}_j \|},$$  \hspace{1cm} (15)

where $\|x\|$ is the norm of the vector $x$, $\varepsilon$ is the machine precision, $n$ is the order of the matrix $A$, $\tilde{\lambda}_j$ and $\tilde{x}_j$ are approximate solutions of Eq. (14). The results of diagonalization procedure DIAGL are considered excellent, if $\tau \leq 1$, good if $1 \leq \tau \leq 100$, and poor if $\tau > 100$. Our tests have shown that the performance index $\tau$ is $10^{-2}$–$10^{-3}$ for $\omega=10^2$–$10^9$ MHz, $F=0.2$–$1000$ kV/cm.

DATA OUTPUT. After completion of diagonalization procedure, we get the wave functions (see Eq. (9)) and the average energies (see Eq. (10)) of an atom in the electric field in the initial coordinate system. Additionally, we have a special file with the expansion coefficients $C_{nk}$ from Eq. (9), where the expansion coefficients are printed in descending order. These data allow the degree of the Stark state interactions to be estimated.

3.2. Part 2 of the package StarkD. Programs for calculating the AC Stark effect and the spectroscopic characteristics of atoms in the electric field.

The obtained output data (the wave functions and the average energies of an atom in the electric field) are further used to calculate the spectroscopic characteristics of the examined atom in the electric field with the parameters $F$ and $\omega$. 
STARK: computes the positions of the Stark components of atomic spectral lines with respect to the position of these lines in the absence of the electric field.

PROBABIL: computes the spontaneous transition probabilities between the Stark states $JM$ and $J'M'$ given by Eq. (11).

INTENS: computes the intensities of the Stark components of spectral lines in different approximations (see Eq. (12) and the discussion after this formula).

The programs STARK, PROBABIL, and INTENS are written in Fortran.

PROFILES: this program calculates the spectral line profiles for the emission spectra of atoms in the electric field using Eq. (13). Spectral lines can be calculated with different profiles, namely: the Lorentz (subroutine LORENTZ), the Doppler (subroutine DOPPLER), and the Voigt (subroutine VOIGT) profiles. The program PROFILES is written in Maple, because this symbolic math package provides the best capabilities for simultaneous calculating and plotting the spectral line profiles.

4. Results and discussion

The package StarkD allows us to study the behavior of the emission spectra of atoms in alternating electric fields with frequency and strength changing in wide ranges. Such wide applicability has enabled us to reveal a number of interesting regularities in the behavior of the emission spectra of rare gas atoms in the electric field. Let us consider the most interesting regularities resulting from our calculations.

1) AC Stark effect. Based on the results of our calculations, we found that the dependence of the Stark component shifts on the electric field strength is quadratic, that is, $\Delta E \sim F^2$, where $\Delta E$ is the Stark component shift with respect to its position in the absence of the electric field. The quadratic dependence is known from perturbation theory, and this fact confirms the validity of our theoretical approach. Additionally, we established that the shifting of the Stark components and splitting of spectral lines in the electric field increase with the principal quantum number $n$ and the orbital quantum number $l$ of the outer-shell electron. Finally, we revealed that an increase in the electric field frequency can lead to the change in the shift directions of atomic spectral lines even under non-resonant excitation by the electric field. As an illustration, Figs. 2 and 3 show such shift-direction changes for the He and Ne atoms.

**Figure 2.** $\omega$-dependence of the shift directions for the $np^3P_0 \rightarrow 2s^3S_1$ spectral lines of the He atom.

**Figure 3.** $\omega$-dependence of the shift directions for the $np' [1/2] \rightarrow 3s' [1/2]$ spectral lines of the Ne atom.

Analogous results are obtained for the emission spectra of Ar and Kr atoms.
2) Transition probabilities. The probabilities of the $J \rightarrow J'$ transitions demonstrate different behavior types for $J < J'$ and $J > J'$. For $J < J'$, switching on the electric field leads to an immediate drop of all probabilities, whereas for $J > J'$, at least one of the probabilities slowly changes with increasing $F$ (see Figs. 4, 5). Additionally, the results of calculations show that switching on the electric field leads to ordering of the transition probabilities with respect to the magnetic quantum number $M$. As can be seen from Fig. 5, for the $J=1 \rightarrow J'=2$ transitions, the probabilities of the $\pm M \rightarrow \max |M'|$ transitions have maximal values, and these probabilities decrease with $M'$. The same regularity is observed for the $J=2 \rightarrow J'=1$ transitions, namely, the probabilities of the $\max |M| \rightarrow \pm M'$ transitions are maximal, and these probabilities diminish with $M$.

These regularities are valid for all examined rare gas atoms, namely: He, Ne, Ar, and Kr atoms.

3) Spectral line intensities. It should be noted that in the cases when the populations of the Stark states are determined by the Boltzmann distribution (thermodynamic equilibrium and LTE plasmas), in the static or dynamic approximations (non-equilibrium plasma) or in the case of equally populated magnetic sublevels (under excitation by the magnetic field with an $X$ line), the behaviour of the Stark component intensities is completely specified by the behaviour of the transition probabilities. Thus, the behaviour of the Stark component intensities in the electric field obeys the same regularities as the transition probabilities. This means that the behaviour of the Stark component intensities differs for $J < J'$ and $J > J'$, and switching on the electric field leads to ordering of the Stark component intensities with respect to the magnetic quantum number $M$. As an illustration of the validity of these conclusions, Fig. 6 demonstrates the behavior of the intensity of different spectral lines of the He atom in the electric field. Analogous results are obtained for the spectral line intensities of Ne, Ar, and Kr emission spectra. The results obtained allow us to understand the reasons for quenching of atomic spectral lines in the electric field.

4) Spectral line profiles. In the framework of our approach, the total profile of atomic spectral line in the electric field is calculated as the sum of the Stark component profiles of this line given by Eq. (13). The Lorentz, Doppler and Voigt profiles can be calculated using programs with the corresponding names (see Fig.1).

To verify the correctness and the efficiency of our theoretical approach, we have calculated the profile of the $2^1P_1-3^1D_2$ spectral line of the helium spectrum emitted by the plasma of a current sheet.
In [17], this profile was observed experimentally in the emission spectrum radiated from the current sheet generated in the plasma excited by the magnetic field with an X line. It was shown in [18] that results of our calculations completely agree with the results [17]. This agreement indicates once more the efficiency of our theoretical approach and the reliability of the package StarkD.

![Intensity vs. Electric Field](image1)

**Figure 6.** He atom. Behavior of intensities $I(JM \rightarrow J'M')$ of the Stark components of spectral lines in the electric field.

The program PROFILES allows us to model the behaviour of the spectral line profiles in the electric field fast and easily. As an illustration, Fig. 7 demonstrates the dependence of the line profiles of the Kr atom on the electric field strength, and Fig. 8 shows changes in the behaviour of the line profiles with an increase in the temperature of the krypton atoms. All calculations of the krypton line profiles excited by the electric field are performed for the Doppler broadened lines. Thus, all total profiles presented in Figs. 7, 8 are the convolutions of the Doppler and AC Stark profiles. The calculation of each case presented in Figs. 7, 8 takes only a few seconds.

![Intensity vs. Electric Field](image2)

**Figure 7.** Kr atom. Evolution of the spectral line profiles in the electric field ($T=30$ eV). Solid line, $F=40$ kV/cm; dashed line, $F=80$ kV/cm; dotted line, $F=120$ kV/cm.

Thus, our program PROFILE is efficient and reliable instrument for the investigation of evolution of the spectral line profiles of atoms in electric fields. The computer simulation of spectral line profiles is very important for plasma diagnostics, because these calculations allow the plasma density and temperature to be determined.
Figure 8. Kr atom. Temperature dependence of the profile of the $6d[1/2]_1 - 5p[1/2]_1$ spectral line.
Solid line, $F=40$ kV/cm; dashed line, $F=80$ kV/cm; dotted line, $F=120$ kV/cm.

If it is necessary, other spectroscopic characteristics of atoms in the electric field, in particular, the lifetimes of atomic energy levels, the oscillator strengths, and so on, also can be calculated using the software package StarkD.

5. Conclusions
It follows from the simulation results that the suggested software package StarkD is an efficient and reliable instrument for the study of the emission spectra of atoms in circularly polarized electric fields with the parameters changing in wide ranges. The results obtained by using this package allow us to analyze mechanisms of the formation of the emission spectra of atoms in the electric field and to explain processes taking place in plasma. Additionally, these results have enabled us to reveal new regularities in the behavior of the spectroscopic characteristics of atoms in the electric field, in particular, the effect of ordering of the transition probabilities and spectral line intensities with respect to the magnetic quantum number at switching on the electric field, and the difference in the behaviour of the transition probabilities and spectral line intensities for the transitions with $J<J'$ and $J>J'$.

Wide potentialities of the suggested package are very useful in solving many practical problems of gas discharge physics, plasma diagnostics, astrophysics and all branches of physics, where one needs to investigate the influence of alternating electric fields on properties of the object of study. Finally, the software package StarkD can be very useful for the development of new radiation sources and for searching the optimal operating mode of already existing radiation sources.

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