

Many-level approximation in calculations of dynamic Stark effect for helium atom

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A new method is proposed for calculation of the dynamic Stark effect in a circularly polarized electric field of arbitrary frequency and intensity. The basic equations of the method are presented. In the framework of the approach proposed, shifts and splits of energy levels for the He atom in the circularly polarized electric field are calculated.

Introduction

The dynamic Stark effect is known to occur as atoms and ions are exposed to the effect of an alternating electric field. This effect essentially consists in the fact that the alternating electric field causes a shift and split of atomic and ion energy levels and formation of the spectrum of quasiharmonics. Besides, resonant population of levels is observed. This process can take place even at a very weak electric field, if its frequency is close to the frequency of some transition.

It seems interesting to study the dependence of the dynamic Stark effect on the frequency and intensity of the external electric field. To achieve this task, it is necessary to solve a nonstationary Schrödinger equation for the system under consideration

$$i \frac{\partial \psi_n(\mathbf{r}, t)}{\partial t} = \hat{H}(\mathbf{r}, t) \psi_n(\mathbf{r}, t), \quad (1)$$

where \hat{H} and ψ_n are the Hamiltonian of the system and the wave function for the n th state of this system, respectively. Equation (1) is solved within the framework of nonstationary perturbation theory.

In the time-dependent field the energy of system is not preserved. Under certain conditions, namely, in the case of a monochromatic external field, we can speak about the Stark shift of energy levels. According to the Floquet theorem, to find the wave function of a system in an external monochromatic field, the sought function in the Schrödinger equation (1) can be presented in the form¹

$$\psi(\mathbf{r}, t) = e^{-i\epsilon t} \varphi(\mathbf{r}, t), \quad (2)$$

where the periodic function $\varphi(\mathbf{r}, t) = \varphi(\mathbf{r}, t + 2\pi/\omega)$ can be expanded in a time Fourier series, so that

$$\psi(\mathbf{r}, t) = e^{-i\epsilon t} \sum_{k=-\infty}^{+\infty} C_k(\mathbf{r}) e^{-ik\omega t}. \quad (3)$$

It can be seen from Eq. (3) that the wave function is a superposition of some stationary states with the energies $\epsilon + k\omega$. This superposition is called a quasi-

energy state, and the parameter ϵ is called the quasi-energy of the system.² The spectrum of $\epsilon + k\omega$ values is referred to as the spectrum of quasiharmonics and is the spectrum of states of the new quantum system of atom + field or, as it is customarily said, dressed atom.

The procedure of solving the Eq. (1) significantly depends on the field polarization (linear, circular, or elliptic), but in any case it faces some problems. First, they are problems connected with the need to fulfill standard restrictions on the applicability of perturbation theory, according to which:

1) the electric field strength should be relatively low, and the perturbation induced by the external electric field should be less than the energy deficit between the neighboring energy levels,

2) to consider the resonant and nonresonant perturbations, it is necessary to use different calculation methods.

Second, depending on whether the excitation by the field is low-frequency or high-frequency, it is also necessary to apply different methods for solving the problem.

Calculation technique

The case of circular polarization of the electric field is the simplest from the viewpoint of theoretical investigation of the dynamic Stark effect. If the field is polarized circularly, the calculations become much simpler, since in this case the spatial and temporal variables can be separated. In the system with the central or axially symmetric Hamiltonian of the unperturbed system, the equation for determination of quasistationary states and quasienergies becomes much simpler, since nonstationary Schrödinger equation (1) is reduced to the stationary one. This simplification is performed using the rotating wave approximation.³

Consider the transition to the rotating coordinate system. In the circularly polarized electric field Schrödinger equation (1) can be written as

$$i \frac{\partial \psi_n(\mathbf{r}, t)}{\partial t} =$$

$$= (\hat{H}_0(\mathbf{r}) - eF(x \cos \omega t \pm y \sin \omega t)) \psi_n(\mathbf{r}, t), \quad (4)$$

where $\hat{H}_0(\mathbf{r})$ is the Hamiltonian of the unperturbed atom; the operator $-eF(x \cos \omega t \pm y \sin \omega t)$ describes the perturbation caused by the interaction of the atom with the electric field having the frequency ω and the strength F , + sign corresponds to the clockwise polarization of the field, while $-$ is for the counterclockwise one. To pass on to the coordinate system rotating about the axis Z with the frequency ω , introduce the following wave function in it

$$\varphi(\mathbf{r}, t) = \exp(i\omega t \hat{J}_z) \psi(\mathbf{r}, t), \quad (5)$$

where \hat{J}_z is the z -component of the atomic operator of total angular momentum. After substitution of the wave function (5) into Eq. (4), we obtain the equation of the form

$$i \frac{\partial \varphi(\mathbf{r}, t)}{\partial t} = \hat{Q} \varphi(\mathbf{r}, t), \quad \hat{Q} = (H_0 - \omega \hat{J}_z \pm Fx). \quad (6)$$

As can be seen from Eq. (6), the operator \hat{Q} is independent of time. Consequently, in the rotating wave approximation, we can pass on from nonstationary Schrödinger equation (4) to the stationary Schrödinger equation of the form

$$\hat{Q} \varphi(\mathbf{r}) = \varepsilon \varphi(\mathbf{r}). \quad (7)$$

The operator \hat{Q} is the operator of quasienergy of the atom in the electric field; ε and

$$\varphi(\mathbf{r}, t) = \exp(-i\varepsilon t) \varphi(\mathbf{r}) \quad (8)$$

are the quasienergy and the wave function of the atom in the electric field in the rotating coordinate system. Obviously, ε and $\varphi(\mathbf{r}, t)$ can be found, using stationary perturbation theory for solution of Eq. (7). Solution of the stationary Schrödinger equation is much simpler than the nonstationary one, but even within stationary perturbation theory some restrictions inherent in it still keep in force.

In the theory of atom/field interactions, numerous attempts have been tried to obtain such equations, which would allow calculation of shifts and splits of atomic energy levels in the field of arbitrary frequency and strength. However, these attempts were successful only for some particular cases. Equations were obtained for calculation of shifts of energy levels of different model systems⁴ or for systems in the single- and two-level approximation.^{5,6} Such equations were obtained for a particle in the short-range potential and for a negative ion (see Refs. 7, 8, and references therein). In Ref. 9 the general equations were derived for calculation of the shift and split of energy levels for an atom in an electric field, but these equations are applicable only in the case that an isolated atomic level is considered in the absence of resonances with the field.

This paper proposes a new method for studying the dynamic Stark effect in a circularly polarized electric field of arbitrary strength and frequency. Within this method, it is suggested to manage without

perturbation theory in solving the stationary Schrödinger equation (7) in order to avoid problems connected with the need to meet the restrictions inherent in the perturbation theory. It is proposed to determine wave functions and energies of the atom in the field that are solutions of stationary Schrödinger equation (7) from diagonalization of the operator of quasienergy \hat{Q} . Write this equation once more for particular n th states of the atom in the electric field with the strength F and frequency ω

$$\hat{Q} \varphi_n(\mathbf{r}) = \varepsilon_n \varphi_n(\mathbf{r}). \quad (9)$$

The wave functions of the operator of quasienergy \hat{Q} will be sought in the form of resolution in terms of the eigenfunctions of the operator $\hat{H}_0(r)$

$$\varphi_n(\mathbf{r}) = \sum_k C_{kn} \varphi_k^{(0)}(\mathbf{r}). \quad (10)$$

Since the wave function (10) is the eigenfunction of the operator \hat{Q} , the matrix of energy of the atom in the field in this representation is diagonal, that is,

$$Q_{mm} = \langle \varphi_m(\mathbf{r}) | \hat{Q} | \varphi_m(\mathbf{r}) \rangle = \varepsilon_n \delta_{mm}, \quad (11)$$

where ε_n is the quasienergy of the atom in the electric field. Therefore, the problem of determining the eigenvalues of the operator \hat{Q} can be considered as a problem of reducing the matrix of this operator to the diagonal form in some representation. In particular, it may be representation of the unperturbed wave functions of the atom or ion as calculated with no external electric field applied. The operator \hat{Q} is an Hermitian self-conjugate operator, so its eigenfunctions and eigenvalues can be always found from diagonalization of its matrix. In the representation of unperturbed wave functions, the matrix elements of the quasienergy operator \hat{Q} have the form

$$Q_{mm} = E_n^{(0)} \delta_{mm} - \omega \langle \varphi_m^{(0)}(\mathbf{r}) | \hat{J}_z | \varphi_n^{(0)}(\mathbf{r}) \rangle \pm F \langle \varphi_m^{(0)} | D_x | \varphi_n^{(0)} \rangle, \quad (12)$$

where $E_n^{(0)}$ is the energy of the n th state of the atom without external electric field; D_x is the x -component of the dipole transition operator.

Diagonalization of the matrix of quasienergy with elements (12) yields a set of quasienergy states, that is, wave functions of the system in the field, and the spectrum of quasienergies n of states of the atom in the field. After diagonalization of the matrix \hat{Q} , we obtain the quasienergies ε_n and the wave functions

$$\varphi_n(\mathbf{r}, t) = e^{-i\varepsilon_n t} \sum_k C_{nk} \varphi_k^{(0)}(\mathbf{r}) \quad (13)$$

for n states of the atom in the external electric field of the frequency ω and strength F in the rotating coordinate system. To find quasienergies of an atom

or ion in the initial coordinate system, it is necessary to perform standard averaging of the energy of the system in the quasienergy state over the period of oscillations. After this averaging, the mean energy of the system in the field in the initial coordinate system is written in the form

$$\begin{aligned} \bar{E}_n &= \langle \psi_n(\mathbf{r}, t) | \hat{H}(t) | \psi_n(\mathbf{r}, t) \rangle = \\ &= \varepsilon_n + \omega \langle \varphi_n(\mathbf{r}) | \hat{J}_z | \varphi_n(\mathbf{r}) \rangle. \end{aligned} \quad (14)$$

It is obvious from Eq. (14) that \bar{E}_n is independent of time, which coincides with the result obtained in Ref. 4.

The wave functions $\varphi_n^{(0)}$ and energies $E_n^{(0)}$ of the unperturbed atom or ion are calculated by either the Hartree–Fock method or the method of nonorthogonal orbitals (in the case that the excited states have lower-lying states of the same symmetry). The equation for calculation of the matrix elements of the dipole moment operator has the form

$$\begin{aligned} \langle \varphi_m^{(0)} | D_x | \varphi_n^{(0)} \rangle &= \langle \gamma JM | D_x | \gamma J' M' \rangle = \frac{(-1)^{J-M}}{\sqrt{2}} \times \\ &\times \left[\begin{pmatrix} J & 1 & J' \\ -M & -1 & M' \end{pmatrix} - \begin{pmatrix} J & 1 & J' \\ -M & 1 & M' \end{pmatrix} \right] \langle \gamma J || D || \gamma J' \rangle, \end{aligned} \quad (15)$$

where the dependence on quantum numbers JM is taken into account in the $3j$ -symbols according to the Wigner–Eckart theorem. For transitions of $l_1^{N_1} l_2 - l_1^{N_1} l_3$ type, the reduced matrix elements $\langle \gamma J || D || \gamma J' \rangle$ are calculated as

$$\langle \gamma J || D || \gamma J' \rangle = Q(\alpha, \alpha', JJ') \langle n_2 l_2 || r || n_3 l_3 \rangle, \quad (16)$$

where $Q(\alpha, \alpha', JJ')$ is the factor depending on the quantum numbers of the calculated transition,

$$\begin{aligned} \langle n_2 l_2 || r || n_3 l_3 \rangle &= \\ &= (-1)^{\frac{l_2+l_3+1}{2}+l_2} \sqrt{\max(l_2, l_3)} \langle R_{n_2 l_2} | r | R_{n_3 l_3} \rangle, \end{aligned} \quad (17)$$

$$\langle R_{n_l} | r | R_{n_l'} \rangle = \int_0^\infty R_{n_l}(r) r R_{n_l'}(r) r^2 dr. \quad (18)$$

Results and discussion

This paper presents some results obtained for He atom in calculating the shifts and splits of the energy levels of this atom under the action of a circularly polarized electric field of different frequencies and the electric field strength. The unperturbed wave functions were calculated by the variational method with nonorthogonal orbitals, and the radial integrals were calculated with the analytical orbitals

$$R_{nl}(r) = C_{nl} \sum_{i=1}^m a_i r^{m_i} e^{-\alpha_i r}$$

by the equation

$$\langle R_{nl} | r | R_{n'l'} \rangle = C_{nl} C_{n'l'} \sum_{i=1}^m \sum_{j=1}^{m'} a_i a_j \frac{(n_i + n_j + 1)!}{(\alpha_i + \alpha_j)^{n_i + n_j + 2}}.$$

The matrix of quasienergy (12) was calculated with the made allowance for 71 energy levels of He atom, and, as a result, 361 magnetic sublevels for He atom in the electric field were obtained. All the calculations were carried out for the LS-coupled He atom.

Figure 1 shows the curves describing the shift of the levels with $J = 0$ depending on the electric field strength F at the field frequency $\omega = 100$ MHz. Since $J = 0$, these levels only shift, but do not split in the electric field.

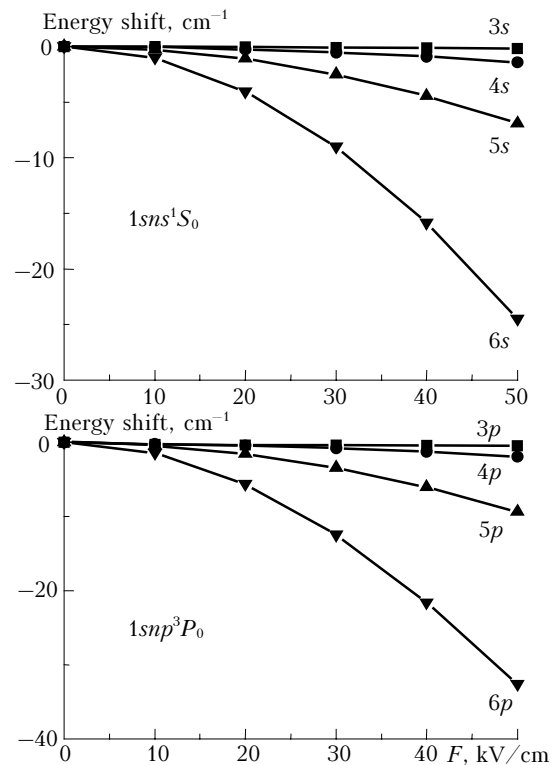


Fig. 1. Relative shift of energy levels with $J = 0$ for He atom as a function of the electric field strength.

As can be seen from Fig. 1, for these levels the square Stark effect is observed, and the higher is the considered level, the larger is the shift, it experiences at the increasing electric field strength.

Now consider the levels that split in the electric field. The plot in Fig. 2 describes the behavior of the $1s2p^3P_1$ and $1s2p^3P_2$ levels of He atom depending on the electric field strength F at the frequency $\omega = 100$ MHz. This excitation is low-frequency and nonresonant (the closest resonance is $\omega = 0.13 \cdot 10^5$ MHz).

As can be seen from Fig. 2, the square Stark effect is observed, and no degeneracy in the magnetic quantum number M takes place. This result agrees with the findings of the perturbation theory. Then, it is seen from Fig. 2 that the split of the levels is small

(about 10^{-4} cm^{-1}) even at a very high field strength $F = 50 \text{ kV/cm}$. The low degree of splitting is explained by the fact that these levels are among the lowest ones, which are low-sensitive to the effect of the electric field.

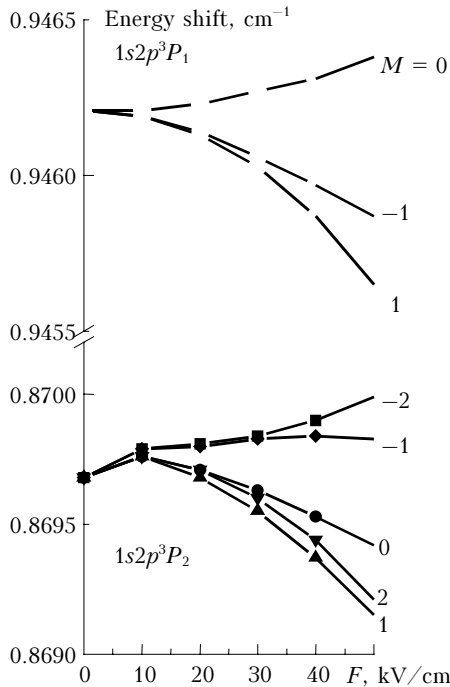


Fig. 2. Split of the $1s2p^3P$ levels of He atom depending on the electric field strength, along with the relative shift ($E_F - E_0 - 169086$), in cm^{-1} .

Consider now the behavior of higher energy levels, for example, $1s4p^1P_1$ and $1s4d^1D_2$ in the electric field with the frequency $\omega = 0.87 \cdot 10^7 \text{ MHz}$. This frequency corresponds to the frequency of the dipole transition $1s4p^1P_1 - 1s4d^1D_2$, so this case is the case of resonant excitation by the electric field. The calculated results are depicted in Fig. 3. It can be seen from Fig. 3 that the degree of splitting of the energy levels in the field increased as compared with the previous result (about 0.5 cm^{-1} at $F = 50 \text{ kV/cm}$). Besides, as in the previous case, the square Stark effect with full relief of the degeneracy in the quantum number M is observed. It should be noted that, unlike perturbation theory, both resonant and nonresonant excitations are calculated uniformly through diagonalization of the matrix \hat{Q} .

Besides, it seems interesting to study how the behavior of the energy levels depends on the frequency of the external electric field. As an example, Fig. 4 depicts the dependence of the $1s6p^1P_1$ level on the electric field strength F at the different frequencies ω . As has already been shown, the higher the energy level, the more sensitive to the effect of the electric field is it. Therefore, to study the behavior of this level, it is sufficient to consider the field strength ranging up to 1 kV/cm , rather than from 0 to 50 kV/cm , as for lower-lying energy levels. As can be seen from the plots depicted, the

split and shift of magnetic sublevels are quite significant (about 0.1 cm^{-1}) even in that weak field.

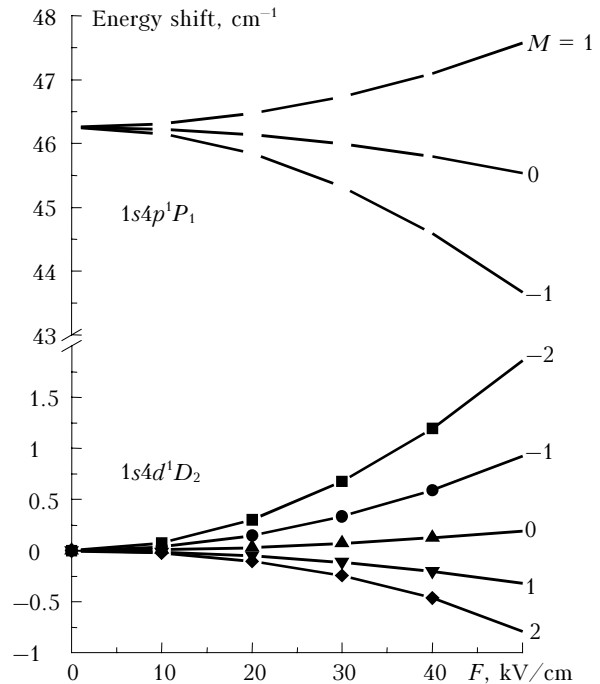


Fig. 3. Relative energy shift of the $1s4p^1P_1$ and $1s4d^1D_2$ levels of the He atom ($E_F - E_0 - 191446.55901$), in cm^{-1} , at resonant excitation by the electric field.

Figure 4a–c demonstrates the behavior of the $1s6p^1P_1$ level in the case of nonresonant excitation by the electric field. It is seen that the sublevel with $M = 0$ is almost insensitive to variation of the field frequency, while the shift of the $M = -1$ sublevel decreases and that of the $M = 1$ sublevel increases with the increasing field frequency. In general, this leads to a situation that at the nonresonant excitation the degree of splitting of the level decreases with the increasing field frequency.

Figure 4d shows the behavior of this level at resonant excitation by the field with $\omega = 0.266 \cdot 10^7 \text{ MHz}$. This frequency is equal to the frequency of the $1s6p^1P_1 - 1s6d^1D_2$ dipole transition. It is seen from Fig. 4d that the degree of splitting of the level decreases (as compared with the previous results) for lower field frequencies, but behavior of the $M = -1$ magnetic sublevel is different than that in the case of nonresonant excitation.

It is interesting to note that the regularity is observed for all the considered levels that under nonresonant excitation by the electric field, the sequence order of the magnetic sublevels is arbitrary, while at the resonance excitation, the magnetic sublevels are ordered by the quantum number M .

The common analysis of the results calculated for He atom reveals some regularities for the dynamic Stark effect in this atom.

1) From analysis of the wave functions it follows that as the electric field strength grows, the degree of interaction among the energy levels increases, while

the growth of the electric field frequency leads to a decrease in the mixing of the levels and in the range of optical frequencies $\sim 10^8$ MHz we deal with almost isolated levels.

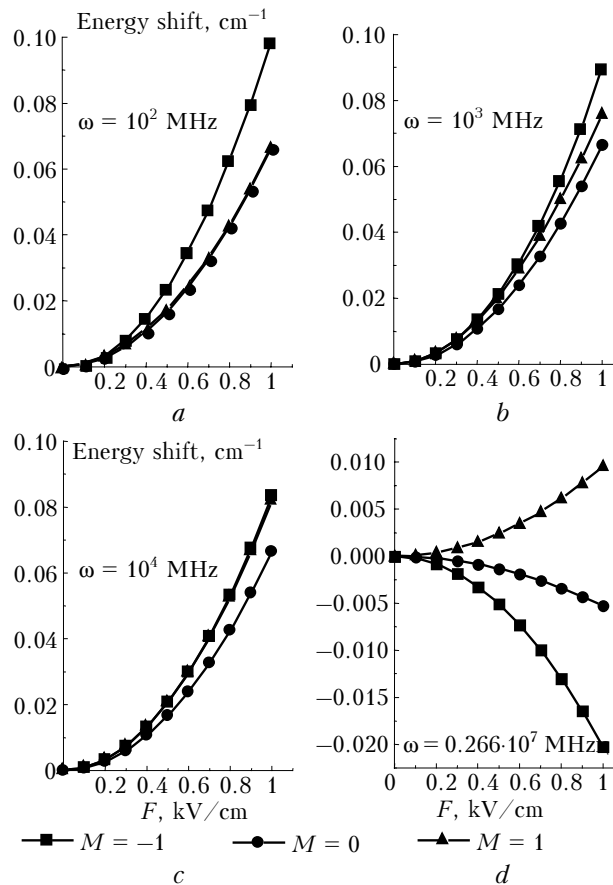


Fig. 4. Behavior of the $1s6p^1P_1$ level of the He atom depending on the frequency of the external electric field.

2) Analysis of the plots describing the shift and split of the energy levels of the He atom in the electric field of different strength and frequency suggests that under the action of a circularly polarized electric field the square Stark effect with the full relief of degeneracy of the energy levels in the magnetic quantum number is observed.

3) The higher the energy level, the larger is the shift, it experiences in the electric field. The increase of the electric field frequency leads to a decrease in splitting of an energy level at the same field strength.

These conclusions agree with the results of perturbation theory, what demonstrates once again the adequacy of the approach proposed.

Conclusions

As can be seen from the above reasoning, the proposed approach to calculation of spectra of atoms

and ions placed in a circularly polarized electric field has the following advantages over the methods of perturbation theory.

1) This approach is free of limitations inherent in perturbation theory and can be applied to calculation of the dynamic Stark effect for any atom or ion exposed to a circularly polarized electric field of arbitrary strength and frequency. Besides, the cases of both resonance and nonresonance excitation are calculated by the same equation and do not require application of various approximations in contrast to the perturbation theory.

2) It seems to be very important that the computational procedure involves many, rather than few energy levels for real atoms and ions. Thus, within the framework of the approach proposed the Stark effect is calculated theoretically in the multi-level approximation, which allows one to additionally estimate the degree of influence of the interaction between atomic levels on the effect under study. Since the energy levels of the atom in the field are very close, the interaction between them should necessarily be taken into account.

3) The proposed approach suits for calculation of both low- and high-frequency excitations.

The method proposed is rather complicated in calculations, but the STARKD program package was developed for its realization. This software makes the calculations much easier.

As can be seen from the results presented, the behavior of spectral lines of atoms and ions in a circularly polarized electric field of arbitrary strength and frequency can be predicted based on calculations within the framework of the method proposed.

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