

SPECTROSCOPY OF ATOMS AND MOLECULES

The Excitation of Rydberg Atoms of Thallium in an Electric Field

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Received May 5, 2017

Abstract—The spectrum of excitation of Rydberg states of thallium atoms has been investigated using a collimated atomic beam in a two-step isotope selective laser scheme $6^2P_{1/2} \rightarrow 6^2D_{3/2} \rightarrow Tl^{**}$ in the presence of an electric field with a strength of up to 1.5 kV/cm near the level $16F_{5/2}$. The optical transitions $6D_{3/2} \rightarrow 18D_{3/2}$ and $6D_{3/2} \rightarrow 16G_{7/2}$, which were induced by an external electric field and dipole-forbidden, have been studied experimentally. The values for the scalar polarizabilities (in units $\text{cm}^{-1}/(\text{kV}/\text{cm})^2$) $\alpha_0(16F_{5/2}) = 3.71 \pm 0.3$, $\alpha_0(18D_{3/2}) = 11.70 \pm 0.25$, and $\alpha_0(16G_{7/2}) = 44.1 \pm 0.9$, which are compared with the calculated one, have been obtained. The new values of energy parameters for the states $18D_{3/2}$ and $16G_{7/2}$ have been determined.

DOI: 10.1134/S0030400X18010046

INTRODUCTION

The quantitative measurements of energy level shifts in atoms, which are caused by the presence of an external electric field, are necessary when testing the accuracy of theoretical methods that are used in the calculation of atomic wave functions in many-electron atomic systems. These calculations are particularly relevant to thallium atoms, which belong to group III of the periodic system. Thallium is an important test system for checking violations of discrete symmetry [1]. The results for thallium can be applied to other trivalent atoms, such as aluminum, gallium and indium. Studies of the Stark effect are of great interest due to its application in the development of a new generation of optical atomic clocks and optical cooling systems, the investigation of long range forces between atomic particles, and so on [2]. These studies can provide information on the electric-dipole moment of the electrons [3] and non-conservation of parity in atoms [4]. Theoretical calculations of electric-dipole polarizabilities for some atoms have developed significantly in recent years [5].

Investigations with Rydberg atoms make up a special group, because in this case the value of the polarizability of atoms in an electric field increases in proportion to the seventh power of the number of the principal quantum number. Accurate experimental data for highly excited states provide a further test of the theoretical calculations. A large number of experiments on Stark spectroscopy were carried out for alkaline and alkaline-earth atoms [6]. However, experimental results for the polarizability of complex atoms are much less reported. This is due to the difficulties in

the experimental realization of the high-resolution spectroscopy of highly excited states.

The thallium atom is one of the heaviest element of group III, which has two stable isotopes ^{203}Tl and ^{205}Tl , with the relative content in the natural mixture, respectively, 29.5 and 70.5% with a spin of 1/2. From a theoretical point of view, the thallium atom has a simple electronic structure with one valent p -electron outside the closed shells, the spectra of which are structured as in hydrogen-like atoms. However, the simple structure of type ns^2nl in Tl is disrupted by the existence of more complex configurations, such as $nsnp^2$. The interaction of these two configurations has a strong influence on various atomic parameters (the intervals of the fine structure, the oscillator strength of electric-dipole transitions, etc.). The quantum defects of the energy states are great as a result of nucleus complex. This leads to a complex picture of the behavior of the spectra of the optical transitions in the presence of electric field. As a result, the Stark structure of thallium energy levels is not similar to the spectra of alkali atoms. The recent calculations of the Stark effect for an atom of gallium, which also belongs to the III group of elements, have shown [7] that its Stark-level structure differs from the structure of alkali metal atoms, which were previously widely studied.

The present article presents the first experimental results on the Stark spectroscopy of Rydberg states of thallium atoms in the vicinity of the level $16^2F_{5/2}$, which were obtained with narrowband frequency-tunable lasers and a collimated atomic beam. The aim of this experimental research was to obtain absolute val-

ues of the scalar polarizabilities of the energy states and their comparison to the calculated ones. In parallel, the forbidden transitions that occur due to the mixing of energy levels with different values of orbital momentum L in the presence of an external electric field were recorded. This fact has been known for a long time. However, it has been theoretically and experimentally poorly explored for complex atoms. Such transitions, which are induced by an electric field for the atoms of group III, were previously examined only for aluminum atoms [8]. This process is of interest for laser separation of the thallium isotopes. In this case, it becomes possible to use narrowband lasers with a fixed radiation frequency for the effective excitation of Rydberg states. Herewith, the precise tuning of the absorption line of the desired isotope to the laser-emission frequency can be performed by changing the strength of the external electric field. This leads to a significant simplification of the route of the selective excitation of atoms for laser isotope separation.

EXPERIMENTAL TECHNIQUE

The full description of the experimental setup and measurement technique is contained in works [9–11]. Only the main details related to the research carried out in this work are presented here. The excitation of Tl atoms to the Rydberg states was carried out in a two-stage scheme $6P_{1/2} \rightarrow 6D_{3/2} \rightarrow nF_{5/2}$. The radiation of the second harmonic of the laser on a Pyrrometene 556 dye ($\lambda = 276.8$ nm, line width $\Delta\nu = 380$ MHz, pulse duration at half-height $\tau \approx 9$ ns) was used in the first stage. In the second stage, frequency-tunable radiation from a second Styryl 11 dye laser ($\lambda = 773$ – 808 nm, $\tau \approx 13$ ns, $\Delta\nu \approx 1$ GHz) was used. The radiation from a copper-vapor laser operating with a repetition rate of $F = 11$ kHz was used as the pump of the dye lasers. The average density of the radiation power of the laser on the first stage was 3 mW/cm², and for the second stage it was 1 W/cm² ($\lambda = 795$ nm) in the absence of absorption saturation for these transitions, which leads to a broadening of the absorption resonance. A collimated beam of thallium atoms with a concentration of $\sim 10^{11}$ cm⁻³ propagated between two rectangular parallel stainless steel plates of a size of 6×15 cm with a gap between them of 2 ± 0.1 cm, where it crossed at a right angle with the combined radiations of the two lasers. One plate was grounded, and the other was under an adjustable pulsed or constant voltage. The interaction region was determined by the transverse size of the laser beam in the area of the atomic beam, which was an ellipse with dimensions of the axes of 1×2 cm and a length of 15 cm. The width of the transition $6P_{1/2} \rightarrow 6D_{3/2}$ in a collimated atomic beam of thallium at the first step narrowed significantly to less than 200 MHz in comparison to the width of the Doppler contour of the absorption ($\Delta\nu_D = 1.7$ GHz) for an atomic vapor, which allows

resolving of the hyperfine and isotopic splitting of the ground state. The frequency of the laser radiation of the second stage with linear polarization, which is parallel to the electric field between the plates, was set on the transition $6^2D_{3/2} \rightarrow n^2F_{5/2}$ and also could smoothly adjust to a wide range of the spectrum.

The populations of the Rydberg states (PRS) $n^2F_{5/2}$ and the others that are studied in this work can be determined by the method of field ionization in a pulsed electric field, which was created between the plates after laser-excitation pulses. At values of the electric field exceeding the critical value for a given state, the ionization of the atom occurs with the efficiency close to 100%. PRS can be determined from the value of the electric current that occurs between the plates. This method has significant drawbacks in experimental studies of the Stark effect. The main one of them is the uncontrollable electric field that arises due to the presence of the residual charge between the plates from a high-voltage ionization pulse at the time ($\tau = 1/F \approx 10^{-4}$ s) of the arrival of the next laser pulse. Since the residual field leads to additional effects in the spectrum of the excitation of Rydberg states, the fluorescence method was used. In this case, the PRS is determined by recording the blue-green fluorescence in a spectral range of 440–460 nm, which arises due to dipole-resolved decays $nP_{1/2, 3/2} - 7S_{1/2}$ of the Rydberg states of Tl atoms. The population of $nP_{1/2, 3/2}$ states after excitation of the $nF_{5/2}$ state is the result of the optical cascade transitions $nF_{5/2} \rightarrow n_1D_{3/2} \rightarrow n_2P_{1/2, 3/2}$. The fluorescence radiation is well recorded visually, and its intensity was measured by using a photomultiplier in a direction perpendicular to the propagation of the atomic beam and laser radiation through a hole with a diameter of 1 cm in the center of one of the plates. The aperture of the hole was closed with a wire mesh to reduce the distortion of the electric field. Filters that shut off the intense ultraviolet radiation of the luminescence and scattered pump radiation at the optical elements of the experimental setup were placed in front of the entrance window of the photomultiplier. The low recording sensitivity associated with the properties of Rydberg states is a disadvantage of this method of determining the PRS. First, the fluorescence intensity decreases rapidly in inverse proportion to the cube of the principal quantum number [12]. The second reason is connected with the large lifetime of the Rydberg states, which exceeds the transit time of the highly excited atom through the observable region [12]. Because of this, the ratio signal/noise for the signal from the photomultiplier output due to excited states $16F_{5/2}$ did not exceed 20. A box-car-integrator, analog-to-digital converter and computer were used to record the absorption spectra. The radiation frequencies of the tunable lasers from the wavelength meter were fixed simultaneously with an accuracy of 300 MHz.

THEORY

The energy state of the level of the thallium atom $W_{n,L,J}$ with principal quantum number n , orbital momentum L , and total momentum J was determined by the formula

$$W_{n,L,J} = I_p - R_y/v_n^2, \quad (1)$$

where $v_n = (n - \Delta_{L,J})$ is the effective principal quantum number of the n -level, I_p is the ionization potential ($I_p = 49\,266.55 \text{ cm}^{-1}$ for the ground state $6^2P_{1/2}$ of the isotope ^{205}Tl with $F=0$ [9]), $R_y = 109\,737.024 \text{ cm}^{-1}$ is the Rydberg constant for the atom of thallium, and $\Delta_{L,J}$ is the quantum defect. The most accurate values known to date of quantum defects $\Delta_{L,J}$ were used for the calculations of the energy of the levels by formula (1). The values $\Delta_F = 1.0344$ were used for the $nF_{5/2}$ states [9]; $\Delta_{p1} = 4.255$ and $\Delta_{p3} = 4.163$ for the $nP_{1/2}$ and $nP_{3/2}$, respectively, states [13]; and $\Delta_S = 4.735$ and $\Delta_D = 3.125$ for states $nS_{1/2}$ and $nD_{3/2}$ [14, 15].

In external electric field E , the shift $\Delta W(n, J, m_J)$ of different magnetic sublevels m_J for a particular electronically excited state can be expressed through the scalar and tensor polarizabilities α_0 and α_2 . If the hyperfine structure is negligible, which is satisfied with good accuracy for the investigated states, then the Stark shift and splitting are described [16] by the following formulas:

$$\begin{aligned} \Delta W(n, J, m_J) &= -\frac{E^2}{2} \left[\alpha_0 + \alpha_2 \frac{3m_J^2 - J(J+1)}{J(2J-1)} \right], \\ \alpha_0 &= -\frac{2}{3(2J+1)} \sum_k \frac{|\langle J|d|J_k\rangle|^2}{W_n - W_k}, \\ \alpha_2 &= 2 \left[\frac{10J(2J-1)}{3(2J+3)(J+1)(2J+1)} \right]^{1/2} \\ &\times \sum_k \frac{|\langle J|d|J_k\rangle|^2}{W_n - W_k} (-1)^{J+J_k+1} \begin{Bmatrix} J & J_k & 1 \\ 1 & 2 & J \end{Bmatrix}. \end{aligned} \quad (2)$$

For these equations, the square of the matrix element $|\langle J|d|J_k\rangle|^2$ for the dipole moment $d = er$ in the J -basis can be computed [16] from the square of the radial-matrix element $|\langle L|d|L_k\rangle|^2$ by the formula

$$\begin{aligned} |\langle J|d|J_k\rangle|^2 &= L_m(2J+1)(2J_k \\ &+ 1) \begin{Bmatrix} L & J & S \\ J_k & L_k & 1 \end{Bmatrix}^2 |\langle L|d|L_k\rangle|^2, \end{aligned} \quad (3)$$

where L_m is the maximum value of L and L_k . The radial-matrix elements of transitions between Rydberg states for of non-hydrogen-like atoms in formula (3)

were determined in the Coulomb approximation by the formulas presented in work [17]:

$$\langle L|d|L_k\rangle = \left\{ \frac{3}{2} v_c^2 [1 - (L_m/v_c)^2]^{1/2} \right\} \sum_{p=0}^3 \gamma^p g_p(s), \quad (4)$$

where $v_c = 2/(1/v_n + 1/v_k)$, $s = v_n - v_k$, $\gamma = \Delta L(L_m/v_c)$, and $\Delta L = L_k - L$. The values for the functions $g_p(s)$ are given in [17]. In [18], it is stated that this method for the calculation of the radial-matrix elements gives an accuracy of less than 0.1% for large values of v_n and v_k . It should be noted that the adjacent transitions with $s \ll 1$ make the main contribution in the calculations of the polarizabilities of Rydberg states. The states with $s \geq 1$ in formula (4) for the dipole moments of the transition yield values three orders of magnitude smaller and, respectively, continue to decrease rapidly with increasing s and ΔL . For the Rydberg states the scalar and tensor polarizations grow as v^7 with increasing principal quantum number [12]. It follows that the contribution of the lower strongly bound state $3D_{3/2}$ to the polarizability can be ignored in the calculations, because it is much less than for the considered upper Rydberg states. The Stark structure in nonhydrogen atoms, even for the simplest alkali metal atoms, is too complex to be fully described by the set of parameters. In most cases, the calculations are limited to constructing an energy-level diagram as a function of the electric-field strength.

Total moment J ceases to be retained in an outer electric field, even a weak one. Only the projection of moment m_J is retained in a uniform field. Therefore, the selection rules for J are not required at radiative transitions even in a weak field and dipole-forbidden lines emerge in the spectrum of atoms. Such forbidden transitions induced by an electric field have been experimentally recorded earlier, since the discovery of the Stark effect. The calculation of the intensities of these lines is reduced to the calculation of the corrections to the wave functions of the stationary states of atoms [19, 20]. Quantitative measurements of the intensity of the forbidden transitions between low-lying states of cesium atoms and the comparison of these results to the calculation were reported in [20]. Similar experiments were carried out in [21], using a more complex system that consisted of a large number of Stark components in a narrow range of energies for the Rydberg states of the xenon atom. Several new features in the behavior of the energies of the levels in an electric field can arise in the Rydberg atoms of thallium with large values of quantum defects for states with high values of orbital moments.

EXPERIMENTAL RESULTS AND DISCUSSION

Only optical transitions to the $16F_{5/2}$ state with the width of the resonance at the second level of 1.1 GHz (curve 1 in Fig. 1) were recorded in the absence of a static electric field in the interaction region of the

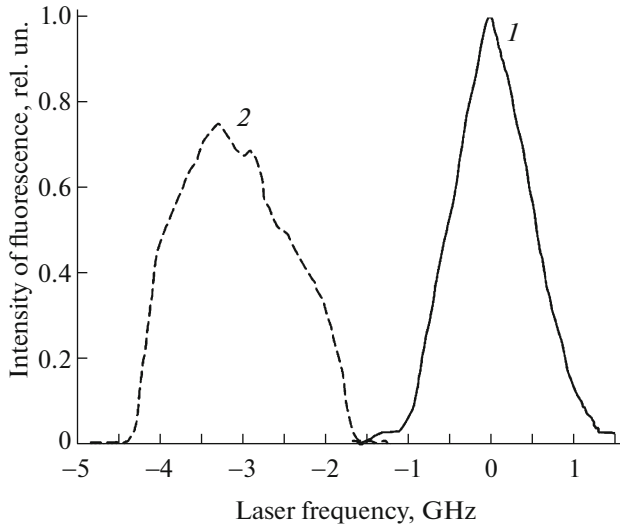


Fig. 1. The spectra of the isotopically selective excitation of a collimated beam of ^{205}Tl atoms on the optical transition of the second stage $6^2D_{3/2} \rightarrow 16^2F_{5/2}$ for two values of the applied electric-field strength: $E = (1) 0$ and $(2) 0.2$ kV/cm.

beam of thallium atoms with laser radiation. At the same time, dipole-allowed transitions to neighboring states $19P_{1/2}$ and $19P_{3/2}$ were not registered experimentally. Oscillator strengths f of these transitions were calculated in the Bates–Damgaard approximation [22] using the tables, which are given in [23]. For the transition $6D_{3/2} \rightarrow 16F_{5/2}$, the calculated value is $f = 1.5 \times 10^{-3}$. For the other optical transitions, $6D_{3/2} \rightarrow 19P_{1/2}$ and $6D_{3/2} \rightarrow 19P_{3/2}$ (the $19P$ states are located in energy close to the $16F_{5/2}$ state), the strength of the oscillators turned out to be 50 and 3000 times less, respectively. It follows that the probabilities of these transitions are small and, therefore, are not observed in the experiment when the signal-to-noise ratio is around ~ 20 for the fluorescence intensity from the $16F_{5/2}$ excited states.

When an electric field was applied, then the center of the resonance, $6D_{3/2} - 16F_{5/2}$ shifts to the long-wave side and its width increases (curve 2 in Fig. 1). A picture of the energy positions of the centers of the optical transitions from the $6D_{3/2}$ to Rydberg states of a ^{205}Tl atom, which were recorded near the level $16F_{5/2}$, depending on the applied electric field is shown in Fig. 2. The value of zero energy on the axis of ordinates coincides with the position of energy $E = 48776.57$ cm^{-1} for the level $16^2F_{5/2}$ ($F = 2.3$), which is counted from the lowest level $6^2P_{1/2}$ ^{205}Tl ($F = 0$). The energy locations of the states that are studied in the work and the values of the fine-splitting were taken from [9, 13–15]. The value of the fine-splitting for F -states in a Tl atom is unknown. The calculations by the formula

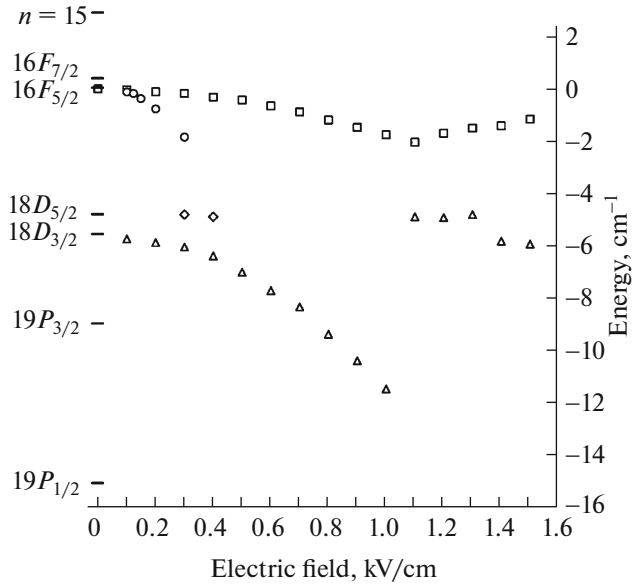


Fig. 2. The energy position of the centers of optical transitions from the $6D_{3/2}$ state to the Rydberg states of a ^{205}Tl atom depending on the applied electric-field strength. The value of zero energy on the axis of ordinates coincides with the position of energy $E = 48776.57$ cm^{-1} for the $16^2F_{5/2}$ ($F = 2.3$) level, which is counted from the lowest level $6^2P_{1/2}$ ^{205}Tl ($F = 0$).

from [24] for the levels $16F_{5/2}$ and $16F_{7/2}$, when compared to the fine-splitting for D -states, show that it is less than 0.36 cm^{-1} . The values of the quantum defects and fine structure for the states $16G_{7/2,9/2}$ are also unknown. When the electric field is $E \leq 1$ kV/cm, the energy shifts of the levels have values smaller than the distance between the levels and, therefore, can be described by the formulas as for the quadratic Stark effect. The huge values of the quantum defects of the energy levels, which are caused by the complicated structure of the atomic nucleus, lead to nonhydrogen behavior of the Stark structure at electric-field strengths of up to 1.5 kV/cm. Thallium is significantly different from the atoms of alkali metals [6] in this respect. The experimental results (curve 2 in Fig. 1) show that the split components in the recorded spectrum are virtually united. This is due to the finite spectral linewidth of the laser radiation at the second level and the presence of inhomogeneity in the applied electric field. Both of them lead to additional broadening of split spectral components m_J .

Since the laser was used in the second level of excitation with linear polarization, which was parallel to the applied electric field, then, according to the selection rules for magnetic quantum number $\Delta m = 0$, only the energy levels with $m_J = 1/2$ and $3/2$ of the $16^2F_{5/2}$, $18^2D_{3/2}$, and $16^2G_{7/2}$ states are excited from the $6D_{3/2}$ state.

It is shown in Fig. 2 that, along with the transition $6D_{3/2} \rightarrow 16F_{5/2}$ with frequency ν_1 , absorption lines on the dipole-forbidden transitions $6D_{3/2} \rightarrow 18D_{3/2,5/2}$ with frequency ν_2 appears in the electric field in the vicinity of the state $16F_{5/2}$. This is due to the mixing of states $16F$ and $18D$, which is caused by an electric field. Accordingly, there are two lines with frequencies ν_1 and ν_2 in the absorption spectrum, which belong to the transitions $6D_{3/2} \rightarrow 16F_{5/2}$ and $6D_{3/2} \rightarrow 18D_{3/2}$, respectively. The number of these forbidden lines is greater in the experimental spectrum, since a larger number of closely spaced states are involved in optical transitions, including the components of the fine structure. The intensity of line ν_1 is practically independent of the field strength, and intensity I_2 of the absorption line with a frequency ν_2 is proportional to the square of the field strength when the dipole moment of the transition is $|\langle 16F|d|18D \rangle| \ll h(\nu_1 - \nu_2)$ in a weak electric field [19]:

$$I_2 = C|\langle 16F|d|18D \rangle|^2/[h(\nu_1 - \nu_2)]^2, \quad (5)$$

where C is a constant. The intensities of lines ν_1 and ν_2 become equal when $|\langle 16F|d|18D \rangle| \gg h(\nu_1 - \nu_2)$ in a strong electric field. The two forbidden transitions $6D_{3/2} - 18D_{3/2}$ and $6D_{3/2} - 18D_{5/2}$ (fine splitting of 0.76 cm^{-1}) are allowed in the presence of an electric field by the selection rules for the matrix element from formula (5). However, calculations by formulas (3)–(5) show that the intensity of the second transition is 14 times less than the intensity of the first one, i.e., that its contribution to the observed pattern of the spectrum is small. In addition, the forbidden transitions to the Stark components $m_j = 1/2$ and $3/2$ for the $16G_{7/2}$ state, which is located above the $16F_{5/2}$ state, are experimentally observed in the presence of an electric field. Therefore, they must be taken into account when calculating the scalar polarization for the $16F_{5/2}$ state.

An ambiguity in the interpretation of experimental results can arise in some cases in the study of optical transitions induced by an electric field. As was noted earlier, it is possible to operate correctly only by the magnitude of the projection of moment m_j for a particular energy state of the atom in the presence of an external, even a weak, electric field. Imagine that the electric field is turned off after the excitation of some Stark component m_j . In which energy state is an atom that is free from the electric field located in this case? The analysis of the field-free Rydberg states of atoms by the method of millimeter-range spectroscopy after the excitation of Stark components m_j and switching off the electric field was carried out using the example of sodium atoms in [25]. The atoms were only in certain energy states, which corresponded to dipole-resolved optical transitions from the initial state, after switching off the field in all cases of the excitation of various Stark components, including for forbidden transitions. Consequently, an excited atom that is in

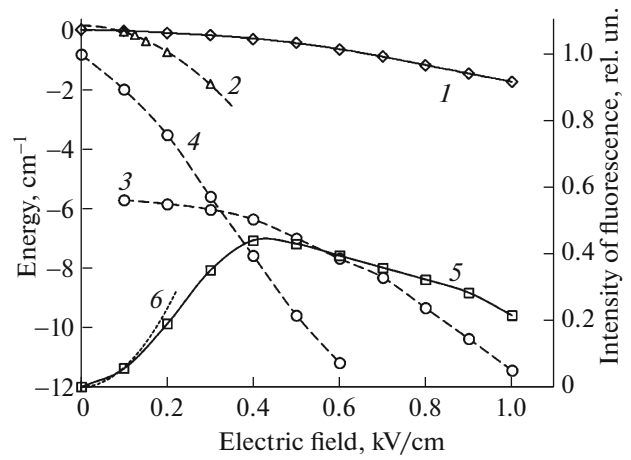


Fig. 3. The energy shift of the levels and the values of the amplitudes of the fluorescence signals for ^{205}Tl in the vicinity of the $16F_{5/2}$ state as a function of the applied electric field. (1–3) The Stark shift of the $16F_{5/2}$, $16G_{7/2}$, and $18D_{3/2}$ states, respectively; (4, 5) the amplitude of the fluorescence signals arising from the decay of the $16F_{5/2}$ and $18D_{3/2}$ states, respectively; and (6) the approximation of the initial section of curve 5 by formula (3).

the electric field when the field is turned off can pass (relax) only in such states that are connected with the initial one through dipole-resolved transitions.

The experimental results for the position of the energy of the levels and amplitude values of the fluorescence signals depending on the applied electric field are presented in Fig. 3. Curves 1–3 represent the Stark shift for the $16F_{5/2}$, $16G_{7/2}$, and $18D_{3/2}$ states, respectively. Curves 4 and 5 show the relative amplitudes of the fluorescence signals that arise from the decay of the $16F_{5/2}$ and $18D_{3/2}$ states, respectively. The amplitude of the fluorescence signal (curve 4) from the decay $16F_{5/2}$ monotonically decreases with increasing applied electric field, although, according to the theory presented in [19], it should not be reduced in a certain initial interval of the electric field. The most likely cause of signal decline is connected with the broadening of the absorption line in the optical transition $6D_{3/2} \rightarrow 16F_{5/2}$ due to the Stark splitting and, as a consequence of this, the broadening of the upper state under monochromatic excitation by laser radiation at the second stage. On the contrary, the amplitude of the signal (curve 5) from the forbidden transition $6D_{3/2} \rightarrow 18D_{3/2}$ first increases with the increasing of the electric-field strength and then falls. The initial growth of the signal amplitude is close to a quadratic dependence, which follows from formula (5) and is illustrated by curve 6 in Fig. 3. The decrease is for the same reason as that for curve 4.

The experimental dependences of the Stark shift of the energy of the levels on the square of the applied electric-field intensity E^2 for the $16^2F_{5/2}$, $18^2D_{3/2}$, and

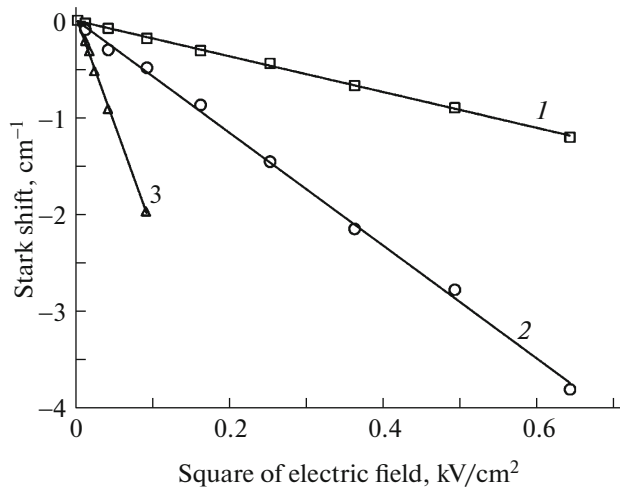


Fig. 4. Dependences of the shift of the energy of the levels on the square of the electric-field strength. (1) The experimental data for the $16^2F_{5/2}$ state and their approximation by the least-squares method by formula (2); (2, 3) the same for the $18^2D_{3/2}$ and $18^2G_{3/2}$ states, respectively.

$16^2G_{7/2}$ states and their approximation by the least-squares method by formula (2) are presented in Fig. 4. The experimental values for the scalar polarizabilities (in units $\text{cm}^{-1}/(\text{kV}/\text{cm})^2$)— $\alpha_0(16F_{5/2}) = 3.71 \pm 0.3$, $\alpha_0(18D_{3/2}) = 11.7 \pm 0.25$, and $\alpha_0(16G_{7/2}) = 44.1 \pm 0.9$ —were obtained from there.

The known data and the results of our measurements were used for the calculations of scalar and tensor polarizabilities of the $18D_{3/2}$ states. In this case, only three neighboring states, $16F_{5/2}$, $19P_{3/2}$, and $19P_{1/2}$, are taken into account (Fig. 2). The contribution of other levels is small compared to the measurement error, as was shown by the analysis. The calculation of the radial-matrix elements by formula (4) for the $18D_{3/2}-16F_{5/2}$, $18D_{3/2}-19P_{3/2}$, and $18D_{3/2}-19P_{1/2}$ transitions showed the values $\langle L|d|L_k \rangle = 328.6$, 326.9 , and 292.8 a.u., respectively. The values of level energy W_0 and quantum defect Δ_D for the $18D_{3/2}$ state were reported earlier in works [14, 15]. Values that are more than an order of magnitude accurate were obtained in the present work by approximating the experimental data in Fig. 3 by the method of least squares according to the formula $W_{18D} = W_0 - bE^2$. The obtained values are $W_0 = -(5.56 \pm 0.04) \text{ cm}^{-1}$ and $\Delta_D = 3.1184$. This measurement accuracy is implemented through the use of narrower absorption resonances and improving of the accuracy of the measurements of the laser-radiation frequencies. The frequency-detuning values ($W_n - W_k$) in formula (2) for the $18D_{3/2}$, $19P_{3/2}$, and $19P_{1/2}$ states, which are equal to 3.43 and 9.52 cm^{-1} , respectively, are taken from the data of [13, 14] and do not exceed the error of their measurement of $<1 \text{ cm}^{-1}$. The calculated value for the scalar polarizability by for-

mula (2) is equal to $\alpha_0(18D_{3/2}) = 6.65 \text{ cm}^{-1}/(\text{kV}/\text{cm})^2$. The discrepancy between theory and experiment is $\sim 43\%$, which is subject to the further refinement of the theoretical model, as all initial parameters are determined with great accuracy. This was to be expected for a complex system that consists of several closely spaced interacting levels. The calculation model does not take into account, for example, the influence of the fine splitting of the $18D_{3/2}$ state, which is comparable to Stark shift of the levels. It can be expected that a refined theoretical model will allow reducing the difference between the calculated values and the experimental ones. The calculation of the tensor polarizability showed that its value is less than 5% of the scalar polarizability. The $18D_{3/2}$ upper level splits only into two symmetrical components, which are simultaneously excited and, therefore, do not contribute an error to the measurement of the scalar polarizability.

It is sufficient to consider only the neighboring $18D_{3/2}$ and $16G_{7/2}$ states in the calculations of the scalar and tensor polarizabilities of the $16F_{5/2}$ level by formula (2). The energy of the $18D_{3/2}$ level and its detuning from the $16F_{5/2}$ state are known with good accuracy. However, the positions of the energy of the G -states of the atom of thallium remain unknown. Therefore, the energy value for the $16G_{7/2}$ state was determined from the assumption that the calculated and measured values for the polarizability coincide. In this case, the best agreement between these values is reached at the detuning of the $16G_{7/2}$ level from the $16F_{5/2}$ one, which is equal to 5.41 cm^{-1} . The values of the radial-matrix element for the transition $16F_{5/2}-16G_{7/2}$ $\langle L|d|L_k \rangle = 320.16$ a.u. and the quantum defect of the state $16G_{7/2}$ $\Delta_G = 0.95$ correspond to this value. The $16F_{5/2}$ upper level splits into three components with $m_J = 5/2, 3/2$, and $1/2$, but only the last two are excited according to the selection rules for magnetic quantum number $\Delta m = 0$ from the $6D_{3/2}$ state. As a result of this splitting, the center of gravity of the total absorption line for the $16F_{5/2}$ state is shifted down in energy by a value that does not exceed 10%. This offset does not exceed the error of measurement of the scalar polarizability of the $16F_{5/2}$ state. Therefore, the splitting of the $16F_{5/2}$ level that is caused by the presence of an electric field has no significant impact on the accuracy of the measurement of the scalar polarizability.

CONCLUSIONS

The Stark effect for the Rydberg states of thallium atoms was studied experimentally in the vicinity of the $16^2F_{5/2}$ level using a collimated atomic beam with a two-stage isotopically selective excitation. The basic atomic data, such as electric polarizabilities and energy parameters of $16F_{5/2}$, $16^2G_{7/2}$, and $18D_{3/2}$ -states, were determined for the first time.

The large density of energy states in a small spectral interval due to large quantum defects and a fine structure leads to spectra that are rather difficult to describe theoretically. However, the peculiarities of the spectra were investigated and comparisons with the calculations in the framework of a simple model were carried out. Strong mixing of energy states with different parities was observed in an electric field experimentally. This makes it possible to excite efficiently thallium atoms with a large concentration through forbidden transitions, adjusting the transition radiation frequency to the frequency of the exciting radiation with the help of a weak electric field.

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Translated by N. Petrov