

Higher-Order Stark Effect on Magnetic Fine Structure of the Helium Atom

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Abstract. We have calculated the scalar and tensor dipole polarizabilities (β) and hyperpolarizabilities (γ) of excited $1s2p\ ^3P_0$, $1s2p\ ^3P_2$ - states of helium. Our theory includes fine structure of triplet sublevels. Semiempirical and accurate electron-correlated wave functions have been used to determine the static values of β and γ . Numerical calculations are carried out using sums of oscillator strengths and, alternatively, with the Green function for the excited valence electron. Specifically, we present results for the integral over the continuum, for second- and fourth-order matrix elements. The corresponding estimations indicate that these corrections are of the order of 23% for the scalar part of polarizability and only of the order of 3% for the tensor part

1 Introduction

The fine structure of the $1s2p\ ^3P$ multiplet of He I has been measured with high precision using both laser spectroscopy [1,2] and microwave techniques [3,4]. These zero-field measurements can be complemented by experiments in non-zero external fields. On the other hand, being the simplest many-body atom, helium has traditionally been the subject of testing the ground of the theoretical approaches to atomic interactions with external fields. Of particular interest are measurements with field strengths, where the energy separation of a pair of sublevels has a minimum. These energy separations at anticrossings are sensitive to matrix elements of the atomic Hamiltonian, which are of no importance in the zero-field fine- structure splitting. Therefore, they provide additional information valuable for detailed tests of the quantum electrodynamical description of the He atom. Energy separations of closest approach have recently been measured for electric-field anticrossings of He I singlet and triplet Stark sublevels [5] and for the $0^- \times 0^-$ anticrossing of the pair of $1s3p\ ^3P_J$ Stark states with $M = 0$ and negative reflection parity at an electric field $F \sim 150$ kV/cm [6]. A corresponding anticrossing occurs also in the Stark splitting of the $1s2p\ ^3P_J$ multiplet at an electric field $F \sim 600$ kV/cm with an energy separation $\Delta E \sim 30$ GHz.

Using modern techniques of laser cooling, high-intensity beams of metastable He ($1s2s\ ^3S$) atoms can be prepared well suited for precision spectroscopy in high electric fields [7]. After transversal cooling, well collimated beams can be exposed

to homogeneous electric fields even as high as 600 kV/cm when traversing a pair of electric-field plates with a spacing of about 1 mm. The energy separation of closest approach of the $0^- \times 0^-$ anticrossing can then be measured with high precision using microwave spectroscopy, since the resonance frequency of the $0^- - 0^-$ transition is independent of the electric field strength in first order and therefore is not broadened by a residual field inhomogeneity. Experimental techniques for detecting these resonance transitions can easily be conceived. For example, one 0^- -sublevel can be excited selectively within the field region using a 1083 nm laser, and the population of the other 0^- sublevel can be selectively detected using two-photon ionization via a resonant intermediate $1s3d\ ^3D$ state.

Using these experimental techniques, the energy separation at the $0^- \times 0^-$ anticrossing can be measured as precise as the zero-field fine-structure splitting. This perspective justifies a thorough theoretical analysis of this anticrossing for determining all energy matrix elements contributing to the energy separation on the ppm level, in order to find out the theoretical parameters, on which the energy separation of the anticrossing depends most sensitively.

In two previous papers [8,9] we have calculated the static polarizabilities and hyperpolarizabilities for $1s3p\ ^3P_J$ ($J = 0, 2$)-states of helium. The method was based on degenerate perturbation-theory expressions for these quantities. The necessary dipole matrix elements were found by using the high-precision wave function on framework of the configuration-interaction (CI) method [10]. The perturbed wave functions are also expanded in a basis of accurate variational eigenstates [11]. These basis sets of the wave functions explicitly take account of electron correlation. To control the result we have also carried out similar calculations with Fues' model potential method.

In this paper we modify and extend this approach in several ways. In particular, we consider the magnetic fine structure effects in the presence of a uniform electric field F for $1s2p\ ^3P_J$ - excited states of helium. We introduce two separate differential polarizabilities to describe the quadratic part of the electric field splitting and three differential hyperpolarizabilities to describe the terms the order of F^4 in the fine-structure splitting of the atomic multiplet $1s2p\ ^3P_J$. We have developed a calculational approach that allows correct estimation of potential contributions due to continuum spectra to the dipole susceptibilities β and γ . In the next section we briefly outline our method. The details of the calculations of the angular and radial matrix elements have been described elsewhere [8,9] and are omitted here for brevity. Atomic units are used throughout.

2 General formulation of method

The Stark effect on the magnetic fine structure occurs as a result of disturbance of atomic levels under the influence of the relativistic and correlation effects as well as the interaction with external electric field F . If the fields are weak enough the centre of multiplet is shifted and there occurs the splitting of sublevels of atomic multiplet n, L, J . The dipole moment induced in an atom by a uniform electric field F is for most purposes expressed as a linear function of F , but higher

terms in F may become important when the field strength becomes sufficiently large. The induced dipole moment D is given by

$$D = \beta F + \frac{1}{6}\gamma F^3 + \dots, \quad (1)$$

in which β is the electric dipole *polarizability*, and γ represents the deviation from a linear polarization law. The parameter γ is the electric dipole *hyperpolarizability*. Such a deviation from linearity corresponds to fourth-order terms in F in the phenomenological expression

$$\Delta E = E - E_0 = -\frac{\beta}{2!}F^2 - \frac{\gamma}{4!}F^4 - \dots, \quad (2)$$

where E_0 is the field-free energy. Both (1) and (2) may be considered as the leading terms in a power series expansion in field strength of the induced moment and induced energy shift, respectively. In the present work we shall show that the Stark splitting of a fine-structure multiplet with states $|nLJM\rangle$ in a field F deviates from the quadratic law in rather weak fields, far from the ionizing one for a given level.

The polarizability in (1) is conveniently expressed in terms of the second-order matrix element of the electric dipole moment component D_z

$$\beta_{nLJM} = 2 \langle \Phi_{nJ}^{(0)} | D_z G_{E_{nJ}} D_z | \Phi_{nJ}^{(0)} \rangle. \quad (3)$$

Here

$$G_{E_{nJ}} = \sum'_{|m\rangle \neq |nJ\rangle} \frac{|\Phi_m^{(0)}\rangle \langle \Phi_m^{(0)}|}{E_m - E_{nJ}} \quad (4)$$

is the reduced Green function of the atom with the energy E_{nJ} , $\Phi_m^{(0)}$ and E_m are eigenstate and energy, respectively, of the zeroth-order Hamiltonian, the symbol \sum' indicating summation over all discrete levels and integrating over the continuum. The corresponding expression for the hyperpolarizability is determined from the Rayleigh-Schrödinger expression for the fourth-order energy

$$\begin{aligned} \gamma_{nLJM} = 24 \left[\langle \Phi_{nJ}^{(0)} | D_z G_{E_{nJ}} D_z G_{E_{nJ}} D_z G_{E_{nJ}} D_z | \Phi_{nJ}^{(0)} \rangle \right. \\ \left. - \frac{1}{2} \beta_{nLJM} S_{nLJM}(-3) \right], \end{aligned} \quad (5)$$

where $S(-3)$ is the sum of oscillator strengths of negative order

$$S_{nLJM}(-3) = \langle \Phi_{nJ}^{(0)} | D_z G_{E_{nJ}}^2 D_z | \Phi_{nJ}^{(0)} \rangle. \quad (6)$$

The polarizability β contains a scalar component β_{nLJ}^s and a tensor component β_{nLJ}^s i.e.

$$\beta_{nLJM} = \beta_{nLJ}^s + \beta_{nLJ}^s \frac{3M^2 - J(J+1)}{J(2J-1)}. \quad (7)$$

The static hyperpolarizability tensor may be resolved into irreducible parts, $\gamma_i(nLJ)$ [8,9] :

$$\begin{aligned} \gamma_{nLJM} = & \gamma_0(nLJ) + \gamma_2(nLJ) \frac{3M^2 - J(J+1)}{J(2J-1)} \\ & + \gamma_4(nLJ) \frac{3(5M^2 - J^2 - 2J)(5M^2 + 1 - J^2) - 10M^2(4M^2 - 1)}{J(2J-1)(2J-2)(2J-3)}. \end{aligned} \quad (8)$$

Here $\gamma_0(nLJ)$ corresponds to the scalar part of hyperpolarizability, $\gamma_2(nLJ)$ corresponds to the rank 2 tensor part of hyperpolarizability, $\gamma_4(nLJ)$ determines the tensor part of the rank 4. As the field strength F increases, the splitting of the level may reach values comparable to the distance between levels of the same parity (the components of the fine structure of an atomic multiplet) and, therefore, the level shift ΔE_{nLJM} can be found by solving the secular equation

$$\det \|\Delta E_{nLJM} \delta_{JJ'} - V_{JJ'}\| = 0. \quad (9)$$

It should be noted, that the off-diagonal matrix element $V_{JJ'}$ is finite only at $J' = J \pm 1, J \pm 2$ and contains a tensor part of polarizability, which depends on the projection M of the total angular momentum J . On the other hand, if we ignore the multiplet splitting in comparison with the energy difference between different multiplets with $n' \neq n$, the matrix element $V_{JJ'}$ can be expressed in terms of the tensor polarizability of the $1sn p^3 P_2$ -state. In our case of $1s2p^3 P_0$ and $1s2p^3 P_2$ states the matrix element V_{02} is given by

$$V_{02} = -\frac{F^2}{\sqrt{2}} \beta_{2^3 P_2}^t. \quad (10)$$

Using (9) and (10) the energy separation $\Delta E_{02}(F)$ in electric field F , may be written in the form

$$\Delta E_{02}(F) = \sqrt{\left[\Delta E_{02}^{(0)} - \frac{1}{2} F^2 \Delta \beta - \frac{1}{24} F^4 \Delta \gamma \right]^2 + 2 F^4 (\beta_{2^3 P_2}^t)^2}, \quad (11)$$

where

$$\Delta \beta = \beta_{2^3 P_0} - \beta_{2^3 P_2}, \quad \Delta \gamma = \gamma_{2^3 P_0} - \gamma_{2^3 P_2}, \quad (12)$$

and $\Delta E_{02}^{(0)}$ is the energy separation of the zeroth-order Hamiltonian. Details of calculation β and γ are presented in the following Sections.

3 Analytical basis set for higher-order calculations of transition amplitudes

The problem of exact *ab initio* calculations of the quantities β and γ is extremely difficult and involves calculating spectral sums over the complete set of unperturbed states. The need for such summation appears in the process of

determining the perturbed wave functions or energy shifts of atomic levels in the perturbation-theory setting. In addition to direct summation over the discrete spectrum and integration over the continuous spectrum of intermediate states, which are extremely involved process in the relativistic case, we basically used two approaches to effectively calculate such spectral sums: a method for integrating inhomogeneous differential equations and a method that uses the formalism of Green functions.

An important advantage of first method is the possibility of using different expressions for the atomic potential, and the calculations can be done not only for a purely Coulomb interaction, but in the multiconfiguration interaction approximation, the Hartree-Fock-Dirac approximation, and the relativistic random phase approximation with exchange effects. The most exact relativistic calculations were done in [12] for the polarizability of the ground state of a helium-like atom.

The effectiveness of the method of Green function is largely determined by the existence of the appropriate representations. Since the analytical representation for the Green function are known only for the Coulomb field, the use of this approach is restricted to problems in which the difference of the potential and the Coulomb potential is insignificant or can be taken into account by perturbation-theory techniques.

In this paper we have taken the Green function for the Fues' model potential from [8,9]. The angular part of $G_E(\mathbf{r}_1, \mathbf{r}_2)$ is simply the product of spherical harmonics, while for the radial part we have taken an expansion in Sturm functions, which have only a discrete spectrum :

$$G_E(\mathbf{r}_1, \mathbf{r}_2) = \sum_{l,m} g_l(E; r_1, r_2) Y_{lm}(\mathbf{n}_1) Y_{lm}^*(\mathbf{n}_2), \quad (13)$$

$$g_l(E; r_1, r_2) = \frac{4Z}{\nu} \sum_{k=0}^{\infty} \frac{U_{kl} \left(\frac{2Zr_1}{\nu} \right) U_{kl} \left(\frac{2Zr_2}{\nu} \right)}{k + \lambda_l + 1 - \nu}, \quad (14)$$

where $\nu = \frac{Z}{\sqrt{-2E}}$, and

$$U_{kl} = \sqrt{\frac{k!}{\Gamma(k+2+2\lambda_l)}} x^{\lambda_l} \exp\left(-\frac{x}{2}\right) L_k^{2\lambda_l+1}(x). \quad (15)$$

The parameter λ_l (l may represent a specific set of spin-orbit quantum numbers, in addition to representing a specific angular momentum) can be found by comparing the quantity

$$E_{nl} = -\frac{Z^2}{2\nu_{nl}^2} \quad (16)$$

with experimental energy values for the lowest state of a valence electron with a given l . The quantity $\nu = \nu_{nl} = n_r + \lambda_l + 1$, with $n_r = 0, 1, 2, \dots$ is the radial quantum number. In particular, for the ground state we have $n_r = 1$ and the effective angular momentum $\lambda = \nu - 2$. Such definition for λ_l implies the existence

of an additional "imaginary" state with the effective principal quantum number $\nu - 1$, since the set of states in the Green function should be complete. On the other hand, the binding energy of the "imaginary" state is almost ten times higher than the excitation energy of any level in a series, so that its contribution to the optical- transition amplitude can be ignored.

The analytical expression for the Green function is particularly convenient when the matrix element under consideration is given by a finite number of the partial terms in expansion (13).

4 Results and discussions

Using the method described in Section 2, we have calculated the scalar and tensor dipole susceptibilities β and γ of excited levels of helium. As noted previously [8,9], the sum in the expression for β is strongly dominated by the term with $nJ = m$ in (4), contributing about 99%. For example, using standard angular momentum theory [13], the resonant term for the scalar polarizability had the form [8,9]:

$$\begin{aligned} \beta_{nLJ}^{s(res)} = & -\frac{2}{3} \sum_{J'} \frac{L(2J'+1)}{E_J - E_{J'}} \left[R_{n,L,J}^{n,L-1,J'} \left\{ \begin{matrix} L & J & 1 \\ J' & L-1 & 1 \end{matrix} \right\} \right]^2 \\ & + \frac{2}{3} \sum_{J'} \frac{(L+1)(2J'+1)}{E_J - E_{J'}} \left[R_{n,L,J}^{n,L+1,J'} \left\{ \begin{matrix} L & J & 1 \\ J' & L+1 & 1 \end{matrix} \right\} \right]^2 \end{aligned} \quad (17)$$

where R are the radial matrix elements, $\left\{ \begin{smallmatrix} a & b & c \\ d & e & f \end{smallmatrix} \right\}$ -is the 6j-symbol [13]. For the circular states where $n = L + 1$, the resonant term in the second part of (17) vanishes. In this case the comparison of equations for β^s and β^t gives $\beta^s \approx -\beta^t$.

The components of the hyperpolarizability have a more complicated resonance dependence. In this connection it is convenient to divide the components of γ into resonant and nonresonant parts: $\gamma_J = \gamma_J^{(res)} + \gamma_J^{(non)}$. The resonant part is determined by the tensor part of multiplet polarizability and the fine-structure splitting $\delta_{JJ'}$ (Table 1).

Because the fine-structure splitting δ_{12} is about one order of magnitude less than δ_{02} , the resonant terms with the factor $1/\delta_{12}$ dominate in these expressions. But they cancel exactly in full expressions for the hyperpolarizabilities of states with the magnetic quantum number $M = 0$. The terms with the factor $1/\delta_{02}$ cancel out of the states with $M = 1$. All resonant terms cancel out of the hyperpolarizability of the state with $M = 2$, because this state has no degeneracy and is not mixed to any sublevel of the n^3P - state.

In the perturbation theory for degenerate states the resonant hyperpolarizability is determined by the tensor part of polarizability [9] and may be extracted out of the fourth-order terms self-consistently: in the case of nondegenerate perturbation theory the resonant part appears for separate sublevels of an atomic multiplet. The numerical results are listed in Table 2.

Table 1. Resonant contributions to the irreducible parts of the hyperpolarizability in the triplet states

J	$\gamma_0^{(res)}$	$\gamma_2^{(res)}$	$\gamma_4^{(res)}$
0	$-12 \frac{(\beta^t)^2}{\delta_{02}^2}$	0	0
1	$-9 \frac{(\beta^t)^2}{\delta_{12}^2}$	$-9 \frac{(\beta^t)^2}{\delta_{12}^2}$	0
2	$\frac{3}{5} (\beta^t)^2 \left[\frac{4}{\delta_{02}} + \frac{9}{\delta_{12}} \right]$	$-\frac{3}{7} (\beta^t)^2 \left[\frac{8}{\delta_{02}} + \frac{9}{\delta_{12}} \right]$	$\frac{18}{35} (\beta^t)^2 \left[\frac{2}{\delta_{02}} - \frac{3}{\delta_{12}} \right]$

Table 2. The numerical values for the components of polarizability and the nonresonant part of hyperpolarizability of helium (a.u.)

Component	Model potential
$\beta_{2^3P_0}^s$	50.69
$\beta_{2^3P_2}^s$	50.73
$\beta_{2^3P_2}^t$	72.12
$\gamma_0^{(non)}(2^3P_0)$	3.673×10^7
$\gamma_0^{(non)}(2^3P_2)$	3.686×10^7
$\gamma_2^{(non)}(2^3P_2)$	3.856×10^6
$\gamma_4^{(non)}(2^3P_2)$	1.077×10^5

Table 3. Contributions of the d -states of the intermediate spectrum to the polarizability of the $1s2p\ ^2P_0$ (a.u.)

n	f_{nd-3p}
3	101.52
4	11.84
5	3.67
6	1.65
7	0.90
8	0.55
9	0.36
10	0.24
Continuum	5.90
Total	126.63

A typical convergence study with the size of the continuum contributions is shown in Table 3 for $1s2p\ ^3P_2$ -state. These corrections are of the order of 23% for the scalar part of polarizability and only of the order of 3% for the tensor part. The magnitude of the electric field and the energy separation at anticrossing $0^- \times 0^-$ are: $F = 632.96$ kV/cm and $\Delta E = 30084$ MHz, respectively.

5 Conclusions

We have calculated the second- and fourth-order dipole susceptibilities of an excited helium atom. Numerical results have been obtained for the $1s2p\ ^3P_0$ - and $1s2p\ ^3P_2$ -states of helium. For the accurate calculations of these quantities we have used the model potential method. The interaction of the helium atoms with the external electric field F is treated as a perturbation to the second- and to the fourth orders. The simple analytical expressions have been derived which can be used to estimate of the second- and higher-order matrix elements. The present set of numerical data, which is based on the Green function method, has smaller estimated uncertainties in ones than previous works. This method is developed to high-order of the perturbation theory and it is shown specifically that the continuum contribution is surprisingly large and corresponds about 23% for the scalar part of polarizability.

It is hoped that this theoretical investigation will add impetus to a the parallel experimental study on atomic characteristics of helium considered.

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