PHYSICAL REVIEW A VOLUME 60, NUMBER 2 AUGUST 1999

Higher-order Stark effect on an excited helium atom

A. Derevianko and W. R. Johnson
Department of Physics, Notre Dame University, Notre Dame, Indiana 46556

V. D. Ovsiannikov
Department of Physics, Voronezh State University, Voronezh 394693, Russia

V. G. Pal'chikov
National Research Institute for Physical-Technical and Radiotechnical Measurements–VNIIFTRI, Mendeleevo, Moscow Region 141570, Russia

D. R. Plante
Department of Mathematics and Computer Science, Stetson University, DeLand, Florida 32720

G. von Oppen
Institut für Atomare und Analytische Physik, Technische Universität Berlin, D-10623, Berlin, Germany

(Received 19 February 1999)

Degenerate perturbation theory is used to study dipole susceptibilities of an excited helium atom in an external electric field. The dependence of the perturbed energy of levels in atoms on fine-structure effects and on the higher-order Stark effect is investigated. Numerical results have been obtained for the $(1s3p)\ ^3P_0$ and $(1s3p)\ ^3P_2$ states of helium. The magnitude of the electric field and the energy separation at the $0^- \times 0^-$ anticrossing are calculated. Calculations of polarizabilities and hyperpolarizabilities are carried out using sums of oscillator strengths and, alternatively, with the excited electron Green function. An estimate is given based on the model potential method for the contribution of an infinite series over the bound states, including the integral over the continuum, for second- and higher-order matrix elements. The relativistic approach for evaluating reduced dipole matrix elements based on the relativistic *no-pair* Hamiltonian and including both the Coulomb and Breit interactions (configuration-interaction method) is analyzed. [S1050-2947(99)06108-9]

PACS number(s): 32.10.Dk, 31.15.Ar, 31.30.Jv, 31.25.Eb

I. INTRODUCTION

Nonlinear susceptibilities of atoms determining higher-order effects in their interaction with strong electromagnetic fields have drawn significant attention in recent years. This interest has been caused both by the development of experimental techniques and by new theoretical approaches for describing higher-order amplitudes of elementary nonlinear processes. The level of experimental accuracy in this field of study is sufficiently high to enable the unequivocal determination of the numerical values for higher-order susceptibilities and for a straightforward comparison between the experimental and theoretical data.

High-resolution spectroscopic measurements on He I atoms in external electric fields may be considered as a new impetus for a detailed theoretical analysis of the state vectors and matrix elements of this fundamental two-electron system. Precision measurements at nonzero electric field can, in particular, be performed at an electric-field anticrossing, where the energy separation of a pair of sublevels has a minimum and is independent of the electric-field strength in first order. In He I, anticrossings of singlet-triplet (S-T) sublevels are found for all $1snl$ levels with $l \geqslant 2$ at electric fields $F \leqslant 100$ kV/cm, which can easily be produced in experiments. These anticrossings arise from the coupling of singlet and triplet states by the spin-orbit interaction H_{SO}. Therefore, the off-diagonal matrix elements $h_{\mathrm{off}} \sim \langle 1snl\ ^1L_L \| H_{\mathrm{SO}} \| 1snl\ ^3L_L \rangle$ can be accurately deduced from these energy separations. So far, precision measurements of S-T anticrossing separations were performed on the $1s3d$ configuration using laser spectroscopy [1] and on the $1s5l$ configurations using level-crossing and radio-frequency spectroscopy [2]. In combination with accurately known zero-field fine-structure splittings, these results can be exploited to determine experimentally all of the relevant parameters of the fine-structure multiplets, i.e., the exchange integrals, the diagonal matrix elements of H_{SS} and of H_{SO}, and the off-diagonal matrix element. Since there are more S-T anticrossings than off-diagonal matrix elements, even higher-order effects may become apparent.

Another type of anticrossing can provide interesting data with respect to relativistic and QED corrections in $1snp$ configurations. This opportunity has been demonstrated by laser spectroscopic investigations of the $0^- \times 0^-$ anticrossing of $(1s3p)\ ^3P$ levels. So far, the energy separation of this anticrossing has been measured only with an accuracy of ± 5 MHz [3]. However, one should bear in mind that here the experimental errors can be reduced by about two orders of magnitude by employing microwave techniques. Nevertheless, even at the present level of accuracy, the measurements make obvious that a simple nonrelativistic approach is insufficient for explaining the experimental result. The ratio of the energy separation at the anticrossing to the zero-field energy separation between the $(1s3p)\ ^3P_0$ and $(1s3p)^3P_2$ levels depends sensitively on relativistic and QED corrections to the scalar susceptibilities of the fine-structure levels. The experimental results of [3] indicate that these corrections are of the order of 1%. After increasing the experimental accuracy by two orders of magnitude, an extremely sensitive test of relativistic and QED corrections will become possible.

Therefore, the time has come for careful relativistic calculations of the susceptibilities for excited states of helium, which involve the most accurate matrix elements, and both fine-structure and mixing effects. In view of the situation presented above, the goal of this paper is a theoretical analysis of the terms up to the order of F^4 in the fine-structure splitting of the atomic multiplet $(1s3p)\,{}^3P_J$ $(J=0,2)$ in a uniform electric field F. We shall show that the fine-structure splitting in a field deviates from the quadratic law in rather weak fields, far from the ionizing one for a given level. Such a deviation corresponds to the fourth-order terms in F in the power-series expansion of the energy shift[1]

$$\Delta E=E-E_0=-\frac{1}{2}\alpha_{nL_{JM}}F^2-\frac{1}{4!}\gamma_{nL_{JM}}F^4-\cdots, \tag{1}$$

where E_0 is the zero-field energy, and $\alpha_{nL_{JM}}$ and $\gamma_{nL_{JM}}$ are the dipole polarizability and hyperpolarizability, respectively. Note that the odd-power terms disappear due to parity conservation. It is well known that the main contribution to the hyperpolarizability $\gamma_{nL_{JM}}$ for fine-structure sublevels is given by the off-diagonal transition matrix element between sublevels, which can be expressed in terms of the tensor polarizability of the atomic multiplet [4,5]. In order to take into account the influence of relativistic effects on this quantity, we use the relativistic configuration-interaction (CI) method, based on the *no-pair* Hamiltonian and including both the Coulomb and Breit interactions [6–8].

In Sec. II, a brief account of our approach is given. The key point is that it is consistent to use relativistic and nonrelativistic variational wave functions for the evaluation of dipole matrix elements, together with experimental transition energies. A description of the CI method and results of full relativistic calculations of reduced dipole matrix elements are given in Sec. III. These results are compared with the values obtained using the Fues model-potential method [10,11] in Sec. IV. A new approach for determining the parameters of the Fues model potential [12] is presented here. Numerical results and discussions are given in Sec. V. Some analytical expressions for the Green function, together with the formulas used for the irreducible parts of the hyperpolarizability tensor are presented in the Appendixes A and B.

II. FORMULATION OF METHOD

The polarizability $\alpha_{nL_{JM}}$ and hyperpolarizability $\gamma_{nL_{JM}}$ in Eq. (1) are conveniently expressed in terms of the second- through fourth-order matrix elements of the atomic dipole moment,

$$\alpha_{nL_{JM}}=2\langle nL_{JM}|DG_{nL_J}D|nL_{JM}\rangle, \tag{2}$$

$$\gamma_{nL_{JM}}=24[\langle nL_{JM}|DG_{nL_J}DG_{nL_J}DG_{nL_J}D|nL_{JM}\rangle -\tfrac{1}{2}\alpha_{nL_{JM}}S_{nL_{JM}}(-3)]. \tag{3}$$

Here

$$S_{nL_{JM}}(-3)=\langle nL_{JM}|D(G_{nL_J})^2D|nL_{JM}\rangle, \tag{4}$$

and

$$G_{(nL_J)}=\sum_{|m\rangle\neq|nL_J\rangle}\frac{|m\rangle\langle m|}{E_m-E_{nL_J}} \tag{5}$$

is the reduced Green function of the atom with the energy E_{nL_J}; the state $|nL_J\rangle$ being excluded from the sum over the complete set $|m\rangle$. The z component of the electric-dipole moment of the atom is given by $D=\Sigma_i r_i\cos\theta_i$ where the summation is carried out over all the atomic electrons.

The structure of $\alpha_{nL_{JM}}$ and relativistic methods for the numerical calculation of the second-order matrix element in Eq. (2) were discussed in detail in our previous paper [12], so we shall focus here on the calculation of the hyperpolarizability given by Eq. (3).

After the angular integration, applying standard angular-momentum theory [13] allows the static hyperpolarizability tensor to be resolved into irreducible parts, $\gamma_j(nL_J)$ [4],

$$\gamma_{nL_{JM}}=\sum_j\left[\frac{(2J+2)_j}{(2J+1-j)_j}\right]^{1/2}C^{JM}_{JMj0}\gamma_j(nL_J). \tag{6}$$

The irreducible parts of the hyperpolarizability may be presented in terms of the fourth-, third-, and second-order reduced matrix elements of the dipole moment,

$$S^{J_1J_2J_3}_{L_1L_2L_3}=\sum_{n_1n_2n_3}\frac{\langle nL_J\|D\|n_1L_{1\,J_1}\rangle\langle n_1L_{1\,J_1}\|D\|n_2L_{2\,J_2}\rangle\langle n_2L_{2\,J_2}\|D\|n_3L_{3\,J_3}\rangle}{(E_{n_1L_{1\,J_1}}-E_{nL_J})(E_{n_2L_{2\,J_2}}-E_{nL_J})}\,\frac{\langle n_3L_{3\,J_3}\|D\|nL_J\rangle}{(E_{n_3L_{3\,J_3}}-E_{nL_J})}$$
$$-\delta_{J_2J}\delta_{L_2L}\sum_{n_1}\frac{\langle nL_J\|D\|n_1L_{1\,J_1}\rangle\langle n_1L_{1\,J_1}\|D\|nL_J\rangle}{E_{n_1L_{1\,J_1}}-E_{nL_J}}\sum_{n_3}\frac{\langle nL_J\|D\|n_3L_{3\,J_3}\rangle\langle n_3L_{3\,J_3}\|D\|nL_J\rangle}{(E_{n_3L_{3\,J_3}}-E_{nL_J})^2}, \tag{7}$$

---

[1] Atomic units are used throughout the paper.

as follows:

$$\gamma_j(nL_J)=24\left[\frac{(2J+1-j)_j}{(2J+1-j)_{j+1}}\right]^{1/2}(-1)^{2J}\Pi_j \times\sum_{j_1j_2}\Pi_{j_1j_2}C^{j_10}_{1010}C^{j_20}_{1010}C^{j0}_{j_10j_20} \times\sum_{J_1J_2J_3}\begin{Bmatrix}1 & 1 & j_1\\ J & J_2 & J_1\end{Bmatrix}\begin{Bmatrix}1 & 1 & j_2\\ J & J_2 & J_3\end{Bmatrix} \times\begin{Bmatrix}j_2 & j_1 & j\\ J & J & J_2\end{Bmatrix}\sum_{L_1L_2L_3}S^{J_1J_2J_3}_{L_1L_2L_3}. \quad (8)$$

Here the standard designations for Clebsch-Gordan coefficients, Pochhammer symbols $(a)_n$, 6-j symbols, and reduced matrix elements of the atomic dipole moment are used [13], and $\Pi_{j_1j_2,\ldots,j_N}=\Pi_{j_1}\Pi_{j_2}\cdots\Pi_{j_N}$, $\Pi_j=\sqrt{2j+1}$. Expressions (7) and (8) can be simplified further for particular sets of atomic quantum numbers J,L.

It should be noted that the general expression for the hyperpolarizability (6) is similar to that of the polarizability, which can be written in terms of the scalar (s) and tensor (t) parts

$$\alpha_{nL_{JM}}=\alpha^s_{nL_J}+\alpha^t_{nL_J}\frac{3M^2-J(J+1)}{J(2J-1)}. \quad (9)$$

The irreducible parts of the hyperpolarizability (8) are nonzero for $j=0,2,4$, and, therefore, the fourth-order term in Eq. (1) is in general determined by only three invariant atomic parameters. The first, $\gamma_0(nL_J)$, corresponds to the scalar part of hyperpolarizability, $\gamma_2(nL_J)$ corresponds to the rank-2 tensor part of hyperpolarizability, $\gamma_4(nL_J)$ determines the tensor part of rank 4.

Finally, the hyperpolarizability $\gamma_{nL_{JM}}$ can be represented in a general form [5]

$$\gamma_{nL_{JM}}=\gamma_0(nL_J)+\gamma_2(nL_J)\frac{3M^2-J(J+1)}{J(2J-1)}+\gamma_4(nL_J)\frac{3(5M^2-J^2-2J)(5M^2+1-J^2)-10M^2(4M^2-1)}{J(2J-1)(2J-2)(2J-3)}. \quad (10)$$

This result may be used directly for determining the energy of a nondegenerate state of an atom. In the case of degenerate states the diagonalization procedure should be used for the atom-field interaction Hamiltonian. Then the above expressions are valid for the diagonal matrix elements of the atom-field interaction, but in this case all the sublevels of the degenerate state should be excluded from the sum for the reduced Green function (5) (see, e.g., [14]).

In view of the goal of this paper, let us consider the energy separation $\delta_{02}=E_{3\,{}^3P_0}-E_{3\,{}^3P_2}$ between fine-structure components with magnetic quantum number $M=0$. Since the zero-field separation $\delta^{(0)}_{02}$ between these components is essentially less than the energy separations between states with different principal and/or orbital quantum numbers, degenerate perturbation theory should be used for determining the energy shift.

Therefore, the energy shift $\Delta E_{nL_{JM}}=E-E_{nL_{JM}}$ is the solution of the secular equation

$$\det\|\Delta E_{nL_{JM}}\delta_{JJ'}-V_{JJ'}\|=0. \quad (11)$$

When determining the energy corrections up to the fourth order in F, we have to calculate the matrix elements in this equation to the same order. The second- and fourth-order corrections to the diagonal matrix elements are determined by the polarizability and hyperpolarizability tensors given by Eqs. (9) and (10). For the off-diagonal matrix element $V_{JJ'}$ which is of second order in F, it is sufficient to take into account only the first nonvanishing term of the corresponding power series because it always appears in the secular equation (11) in a product with another nondiagonal matrix element. $V_{JJ'}$ can be expressed in terms of the tensor polarizability of the $1snp\,{}^3P_2$ state [4,12]

$$V_{02}=-\frac{F^2}{\sqrt{2}}\alpha^t_{n^3P_2}. \quad (12)$$

We now substitute the expressions for the matrix elements into Eq. (11) to obtain the corresponding power series for the energy separation $\delta_{02}(F)$ in a uniform electric field. Since the degenerate basis consists of only two states, this equation is easily solved analytically, and it is possible to calculate the leading terms in the F power series:

$$\delta_{02}(F)=\sqrt{\left[\delta^{(0)}_{02}-\frac{1}{2}F^2(\alpha^s_{n^3P_0}-\alpha^s_{n^3P_2})-\frac{1}{24}F^4(\gamma_{n^3P_0}-\gamma_{n^3P_2})\right]^2+2F^4(\alpha^t_{n^3P_2})^2}\approx\delta^{(0)}_{02}-\frac{1}{2}F^2(\alpha^s_{n^3P_0}-\alpha^s_{n^3P_2}+\alpha^t_{n^3P_2}) -\frac{1}{24}F^4[\gamma_0(n^3P_0)-\gamma_0(n^3P_2)+\gamma_2(n^3P_2)-6\gamma_4(n^3P_2)]+F^4\frac{(\alpha^t_{n^3P_2})^2}{\delta^{(0)}_{02}}+\cdots. \quad (13)$$

The second expression here appears as a power-series resolution for the square root in the first line. It coincides with the field-dependent splitting of the fine-structure sublevels as calculated in nondegenerate perturbation theory which may be used when the field-dependent contribution is much smaller than the field-free splitting $\delta_{02}^{(0)}$. The last term determines the main contribution to the difference between the sublevel hyperpolarizabilities due to the largest contribution (with $n_2=n$ and $L_2=L$, hence, with the smallest denominator) in the sum over n_2 of Eq. (7). Evidently, as noted earlier [4,5,12], this term (we shall call it ''the resonant term'') appears already in the second-order perturbation theory for degenerate states. The previous term, also of fourth order in F, appears only in fourth-order perturbation theory and accounts for the ''nonresonant'' contributions to the hyperpolarizability (with $n_2\neq n$ and/or $L_2\neq L$). However, this term may provide a contribution at the level of a few percent to the total fourth-order correction, which amounts in the vicinity of the anticrossing to a half of the total field-dependent part of the splitting (13). Thus, the contribution of the ''nonresonant'' terms of the hyperpolarizability may be approximately equal to that caused by the relativistic corrections to the polarizability and may be rather important when comparing the theoretical results with high-precision experimental data.

Accurate calculations of the reduced matrix elements for the components of the polarizability and hyperpolarizability require reliable wave functions which should give correct values for both of the quantities. The calculations of the atomic susceptibilities were carried out using a sum of oscillator strengths. To include relativistic effects, we used precise relativistic configuration-interaction calculations of reduced dipole matrix elements (see the next section). To control the results we have also carried out similar calculations with the Fues model-potential method. A simple analytical presentation for the model-potential Green function enables us to evaluate the contribution of the high-*n* terms (including continuum) which are neglected in the finite-sum approach. Details of the calculations and numerical results are presented in the following sections.

III. RELATIVISTIC CONFIGURATION-INTERACTION CALCULATIONS OF REDUCED DIPOLE MATRIX ELEMENTS

The configuration-interaction method has been previously used to obtain high-precision wave functions and matrix elements of atoms with a few electrons, especially for heliumlike ions. In this section we follow the techniques of Ref. [6] to calculate the reduced matrix elements for resonant terms in the polarizability and hyperpolarizability expansions. The wave functions for the atomic states are obtained from relativistic CI calculations. For the initial (final) state, we write

$$\Psi_{I(F)}=\sum_{k\geqslant l} C_{kl}^{I(F)}\Phi_{kl}, \tag{14}$$

where Φ_{kl} are configuration state functions coupled to given values of J,M, and parity, defined by

$$\Phi_{kl}=\eta_{kl}\sum_{m_k m_l} C^{JM}_{j_k m_k j_l m_l}\, a_k^\dagger a_l^\dagger|0\rangle. \tag{15}$$

Here $\eta_{kl}=1/\sqrt{2}$ if $k=l$ and $\eta_{kl}=1$, otherwise. The coefficients $C_{kl}^{I(F)}$ are configuration weights for the initial (final) state determined variationally, seeking extrema of the expectation value of the *no-pair* Hamiltonian including both the Coulomb and Breit interactions. The wave function is subject to the normalization condition

$$\sum_{l\geqslant k}|C_{kl}^{I(F)}|^2=1. \tag{16}$$

Discussions of the relativistic CI problem for heliumlike ions including extensive comparisons of CI energies with experiment have been given in Refs. [7,8]. The electric-dipole operator Q_M in second-quantized form is given by

$$Q_M=\sum_{ij}(r_M)_{ij}a_i^\dagger a_j, \tag{17}$$

where r_M is a one-particle dipole matrix element of the Mth component of the coordinate vector in a spherical basis. By calculating the matrix element of the many-body electric-dipole operator in the CI basis and performing the angular reduction, one obtains an explicit expression for the reduced matrix elements in terms of the weight coefficients $C_{kl}^{I(F)}$ given by Eq. (44) of Ref. [6]. Of particular importance in calculating reduced matrix elements relativistically is the inclusion of contributions from the negative-energy states. As our calculations are performed using the *no-pair* Hamiltonian, contributions from these states are naturally not included and must be added in separately. In Ref. [6], the importance of these contributions was addressed in detail by using second-order QED perturbation theory with the full Hamiltonian (rather than the *no-pair* Hamiltonian used for the present calculations). While contributions from negative-energy states were found to play an important role in the velocity gauge for electric-dipole matrix elements, such contributions in the length gauge were insignificant. As discussed in [9], the relative contribution of negative-energy states for allowed E1 transition amplitudes calculated in length gauge is of order α^4Z^3. As a result of such analysis, our calculations are presented in the length gauge. As a first step in our calculation, we obtain the CI wave functions for the initial and final states. The numerical methods used to evaluate the weight coefficients $C_{kl}^{I(F)}$ are described in Refs. [7,8]. The single-particle basis orbitals used in the CI expansion consist of a subset of 20 out of 30 *B*-spline basis functions for each partial wave. The accuracy of matrix elements was tested by slightly changing the cavity radius and number of included splines. The numerical accuracy achieved is at the level of number of quoted figures in Table I. The conventional approach to CI calculations is to perform a sequence of calculations with increasing number of partial waves included and then to extrapolate the sequence of values to a number corresponding to an infinite set. In the particular case of dipole matrix elements we find that this sequence of values saturates very early and, hence, the results for a set which includes the s, p, d, f, and g partial waves are found to be sufficient.

Finally, in Table I we present the calculated values of reduced dipole matrix elements relevant to resonant contributions to the polarizabilities and hyperpolarizabilities for

TABLE I. Reduced matrix elements of electric-dipole operator (a.u.) for He I.

Transition $|n'L'_{J'}\rangle \rightarrow |nL_J\rangle$	Value $\langle n'L'_{J'}\|D\|nL_J\rangle$
$1s3p\ {}^3P_0 \rightarrow 1s3s\ {}^3S_1$	-6.4797
$1s3p\ {}^3P_2 \rightarrow 1s3s\ {}^3S_1$	-14.489
$1s3p\ {}^3P_0 \rightarrow 1s3d\ {}^3D_1$	8.2923
$1s3p\ {}^3P_2 \rightarrow 1s3d\ {}^3D_1$	1.8542
$1s3p\ {}^3P_2 \rightarrow 1s3d\ {}^3D_2$	7.1805
$1s3p\ {}^3P_2 \rightarrow 1s3d\ {}^3D_3$	16.994
$1s4f\ {}^3F_2 \rightarrow 1s3d\ {}^3D_1$	-13.68
$1s4f\ {}^3F_3 \rightarrow 1s3d\ {}^3D_2$	-16.66
$1s4f\ {}^3F_2 \rightarrow 1s3d\ {}^3D_2$	-5.887
$1s4f\ {}^3F_3 \rightarrow 1s3d\ {}^3D_3$	-5.886
$1s4f\ {}^3F_2 \rightarrow 1s3d\ {}^3D_3$	-0.9953
$1s4f\ {}^3F_4 \rightarrow 1s3d\ {}^3D_3$	-20.03

the cases considered in this paper. The signs of the matrix elements were set consistently with the phase convention used in Fues model-potential calculations, discussed in the next section.

IV. FUES'S MODEL-POTENTIAL METHOD IN CALCULATIONS OF OPTICAL TRANSITION AMPLITUDES IN ATOMS

The Fues model potential (FMP) for describing the states of a valence electron in a many-electron atom is a rather useful tool for simple and straightforward calculations of the optical transition amplitudes in any order of interaction between an atom and an external electromagnetic field. It was proposed by Simons [10] as an analytically solvable example of an atomic pseudopotential which may be used in generating valence and Rydberg orbitals for calculating oscillator strengths and ionization amplitudes. Later on, an analytical expression for an atomic Green's function [11] was derived with the help of the FMP which was quite effective in calculating numerous higher-order transition amplitudes and electromagnetic susceptibilities determining various processes of interaction between atoms and intense fields [5]. Being of an asymptotic nature relative to the valence-electron position vector **r**, the FMP provided accurate numerical results for the matrix elements of positive powers of the absolute value of **r**, specifically for atoms with a single valence electron or the one-electron excited states of an atom with a few valence electrons.

However, it was noted that the method, while giving quite accurate results for excited states, failed to do so for the transitions involving the ground-state wave functions, even in atoms with a single electron in the valence shell or for those involving the metastable states of atoms with several valence electrons. In these cases the wave function of another asymptotic method—the quantum defect method (QDM)—was much more precise in numerical calculations [10,11]. We propose here an approach for determining the parameters of the FMP providing accurate numerical results in calculating static optical characteristics without resorting to any other kind of wave function for the states of atom.

The FMP method, similarly to the QDM, is based on the Rydberg formula for the relation between the energy and the effective principal quantum number ν_{nl} of an atomic level $|nl\rangle$, that is,

$$E_{nl} = -\frac{Z^2}{2\nu_{nl}^2}, \tag{18}$$

where Z is the charge of the residual ion. Here, the notation l is used for the valence-electron angular momentum, in contrast with the total atomic angular momentum L. Therefore $L=l$ if the angular momentum of the atomic core is zero. The corresponding radial wave function of the state has the hydrogenlike form,

$$R_{nl} = (-1)^k \frac{2}{\nu_{nl}^2}\sqrt{\frac{Z^3\ n_r!}{\Gamma(\nu_{nl}+\lambda_l+1)}}\left(\frac{2Zr}{\nu_{nl}}\right)^{\lambda_l} \times e^{-Zr/\nu_{nl}} L_{n_r}^{2\lambda_l+1}\left(\frac{2Zr}{\nu_{nl}}\right), \tag{19}$$

and depends on the choice of the radial quantum number n_r, which in turn determines the effective angular-momentum quantum number, $\lambda_l = \nu_{nl} - n_r - 1$ (a smooth dependence of λ_l on energy of the level in most cases may be neglected).

If we require the experimentally observed set of valence-electron states with angular momentum l to be complete, the value of $n_r = 0$ must be taken for the lowest of them [10,11], consecutively increasing by unity for higher states. This definition caused substantial inaccuracy for the radial matrix elements $\langle nl|r^L|n'l'\rangle$ with $L \geqslant 1$ (up to 50% in absolute value, with an opposite sign in some cases, since the phase-factor integer power k was taken always to be zero). That is why the nodeless ground-state radial wave functions (19) were usually replaced by the one-node QDM wave functions [10,11]. Such a replacement is equivalent to another definition of the radial quantum number, i.e., $n_r = 1$ for the ground state and the effective angular momentum $\lambda = \nu_g - 2$, instead of $\lambda = \nu_g - 1$, where ν_g is the effective principal quantum number of the ground state. Such a modification of the definition for λ implies the existence of an additional ''imaginary'' state with the effective principal quantum number $\nu_{\mathrm{im}} = \nu_g - 1$, since the set of states (19) should be complete. But being of a very low energy, $E_{\mathrm{im}} = -Z^2/(2\nu_{\mathrm{im}}^2)$, and, consequently, having a very short-ranged wave function, the ''imaginary'' state cannot provide significant contributions to the optical transition amplitudes in higher orders.

The modified definition of the radial and effective angular-momentum quantum numbers seems preferable when using the FMP in numerical calculations involving the ground, metastable, and possibly some low-lying states with small angular momenta. The value of $k = n - l - n_r - 1$ should be taken in the phase factor $(-1)^k$, in accordance with the asymptotic phase of the corresponding hydrogenlike wave function.

Table II lists the results of numerical calculations of the ''oscillator strengths,'' $f_{n'S,3P}(-2) = 2|\langle n'\ {}^3S_1|d_z|3\ {}^3P_0\rangle|^2/(E_{n'S} - E_{3P})$ corresponding to the dipole transitions from the first nine $n\ {}^3S_1$ states to the $3\ {}^3P_0$ state in helium which are required to determine the higher-order matrix elements for the static polarizability and hyper-

TABLE II. The contributions of $1sn's\ ^3S_1$ intermediate states on the polarizability of $1s3p\ ^3P_0$ state of He I [11].

n'	$f_{n'S,3P}(-2)\vert_{\lambda_0=0.698}$	$f_{n'S,3P}(-2)\vert_{\lambda_0=-0.302}$	$f_{n'S,3P}(-2)\vert_{\mathrm{CI,var.meth}}$
2	−0.34	−1.67	−1.57
3	−2377.93	−2682.56	−2638.45
4	451.87	319.37	312.17
5	25.38	18.27	17.92
6	6.36	4.60	4.72
7	2.59	1.88	1.85
8	1.34	0.97	0.96
9	0.79	0.57	0.57
10	0.51	0.37	0.34
$\Sigma_{n'}$	−1888.6	−2337.7	−2301.50

polarizability of the 3 $^3P_0$ state. The principal quantum numbers of the ^3S states are given in the first column. The second column presents the oscillator strengths as calculated in the FMP approximation with the usual choice of the radial quantum number $n_r=0$ for the lowest triplet state of the He atom, 2 $^3S_1$, giving the value of the effective angular-momentum quantum number for the ^3S states $\lambda_0=0.698$. The third column corresponds to the modified definition of $n_r=1$ for the 2 $^3S_1$ state, with $\lambda_0=-0.302$. High-precision variational values for the oscillator strengths [13] and numerical data of the CI method [6] for $n'=3$ are presented for comparison in the fourth column. The result of summation over n' is also given in the bottom line of the table. The advantage of the modified definition of the parameters for the FMP wave function is evident (less than 1.6% discrepancy against more than 20% of the usual approach) and may be much more significant for higher-order amplitudes.

We have also computed in the FMP approximation the matrix elements of Table II. They differ in magnitude from the data of Table II by less than 1%, if the modified definition of the angular momentum is used, while for the traditional definition this difference is as much as 20%.

The main difficulty in the exact computation of the amplitudes of second to fourth orders in Eqs. (2) and (3) is related to the infinite sums over the total atomic spectrum including the continuum, which represent the Green function (5). Therefore, for practical numerical computer implementation of the perturbation theory it is necessary to have the most convenient representation for the Green function. In our calculations we used the analytical representation of the Green function for the Fues model potential (Appendix A) based on the Sturm-series expansion [11]. Thus, the radial matrix elements, to which the polarizabilities and hyperpolarizabilities may finally be reduced, can be presented in the form of absolutely converging series, well suited for the numerical computations.

V. RESULTS AND DISCUSSION

Accurate *ab initio* calculations of the $\alpha_{nL_{JM}}$ and $\gamma_{nL_{JM}}$ are not routine because both relativistic and electron correlation effects can be very important. Note that in Eqs. (4) and (5), the transition energies appear in the denominator. Since the resonance energies with $n'=n$ are typically two orders of magnitude smaller than the nonresonant ones $(n'\neq n)$, the resonance terms totally dominate the sum. For these transitions, we used precise relativistic CI calculations of reduced dipole matrix elements. The matrix elements for the other exited states are calculated by using nonrelativistic variational data [13]. The convergence of the method is considered in [12].

Using this approach, we have calculated relativistically the scalar and tensor polarizabilities of helium excited states. For example, the resonant term for the scalar polarizability had the form

$$\alpha^{s(r)}_{nLJ}=-\frac{2}{3}\Bigg\{\sum_{J'}\frac{L(2J'+1)}{E_{nL_J}-E_{nL-1_{J'}}}\left[R^{nJ'L-1}_{nJL}\begin{Bmatrix} L & J & 1\\ J' & L-1 & 1\end{Bmatrix}\right]^2$$

$$+\sum_{J'}\frac{(L+1)(2J'+1)}{E_{nL_J}-E_{nL+1_{J'}}}\left[R^{nJ'L+1}_{nJL}\begin{Bmatrix} L & J & 1\\ J' & L+1 & 1\end{Bmatrix}\right]^2\Bigg\},\tag{20}$$

where $R^{n'J'L'}_{nJL}=\langle n'J'L'|r|nJL\rangle$ are the radial matrix elements. The singlet-triplet mixing corrections between states with the same n,L, and J in Eq. (20) are considered in [11,12]. The numerical values for the mixing parameter

$$\beta_{nL_L}=\begin{cases}\cos(\theta_{nL}), & J=L\\ 1, & J\neq L\end{cases}\tag{21}$$

are listed in [12].

As seen from Eqs. (7) and (8), the components of the hyperpolarizability have a more complicated resonance dependence. Therefore, it is convenient to divide the components (8) into resonant and nonresonant parts: $\gamma_j=\gamma_j^{(\mathrm{res})}+\gamma_j^{(\mathrm{non})}$. As was already pointed out, the resonant part $\gamma_j^{(\mathrm{res})}$ is determined by the tensor part $\alpha^t_{nL_J}$ of the multiplet polarizability and the fine-structure splitting $\delta_{JJ'}$. Such resonant contributions to the irreducible parts of the hyperpolarizability are given in Table III for an $n\ ^3P_J$ state. Generally speaking, since 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 $M=0$, as may be verified from Eq. (10). It is easy to see also that the terms with the factor $1/\delta_{02}$ cancel out of the hyperpolarizability of states with $M=1$. And at last, all the resonant

TABLE III. Resonant contributions to the irreducible parts of the hyperpolarizability in $n\,^3P_J$ states.

J	$\gamma_0^{(\mathrm{res})}(n\,^3P_J)$	$\gamma_2^{(\mathrm{res})}(n\,^3P_J)$	$\gamma_4^{(\mathrm{res})}(n\,^3P_J)$
0	$-12\dfrac{(\alpha^t_{n\,^3P_2})^2}{\delta_{02}}$	0	0
1	$-9\dfrac{(\alpha^t_{n\,^3P_2})^2}{\delta_{12}}$	$-9\dfrac{(\alpha^t_{n\,^3P_2})^2}{2\delta_{12}}$	0
2	$\frac{3}{5}(\alpha^t_{n\,^3P_2})^2\left[\dfrac{4}{\delta_{02}}+\dfrac{9}{\delta_{12}}\right]$	$-\frac{3}{7}(\alpha^t_{n\,^3P_2})^2\left[\dfrac{8}{\delta_{02}}+\dfrac{9}{\delta_{12}}\right]$	$\frac{18}{35}(\alpha^t_{n\,^3P_2})^2\left[\dfrac{2}{\delta_{02}}-\dfrac{3}{\delta_{12}}\right]$

terms cancel out of the hyperpolarizability of the state with $M=2$, i.e., $\gamma^{(\mathrm{res})}_{n\,^3P_{22}}=0$, in complete accord with the fact that this state has no degeneracy and is not mixed to any other sublevel of the $n\,^3P$ state.

Note that the resonant part appears in the hyperpolarizability if we use the nondegenerate perturbation theory for separate sublevels of an atomic multiplet. In this case the resonance terms are retained in the sums over the atomic basis [the sum over n_2 of Eq. (7) or the sum for the Green function (5)].

In the perturbation theory for degenerate states [13], which deals with the secular equation (11), the resonant hyperpolarizability is extracted out of the fourth-order terms self-consistently. Thus the resonant hyperpolarizability is presented explicitly by the last term of Eq. (13) while the rest of the γ components in this equation include only the nonresonant part.

To calculate the nonresonant hyperpolarizability components, $\gamma_j^{(\mathrm{non})}$, we have to remove from the series (7) [or alternatively, from the Green function (5)] all the degenerate substates and to carry out the summation over the rest of the atomic basis. Some of the calculations may be carried out analytically with the use of the angular-momentum algebra, reducing the right-hand side of Eq. (8) to a combination of the radial matrix elements.

We take into account the fact that the nonresonant contribution to the hyperpolarizability is not more than 2–3 % of the total value of γ. So the relativistic effects may be neglected in their calculations. In this case the radial matrix element $\langle n_1J_1L_1|r|n_2J_2L_2\rangle$, which represents the reduced dipole matrix element between triplet states according to the relation [15]

$$\langle n_1J_1L_1\|D\|n_2J_2L_2\rangle=(-1)^{J_2+L_1}\Pi_{J_1J_2L_2}C^{L_10}_{L_2010}\times\begin{Bmatrix}1 & L_1 & J_1\\ 1 & J_2 & L_2\end{Bmatrix}\langle n_1J_1L_1|r|n_2J_2L_2\rangle, \tag{22}$$

may be considered to be independent of the total momenta, and the sum over J_1,J_2,J_3 in Eq. (8) may be calculated analytically. Since the Kronecker symbols of Eq. (7) eliminate the sum over J_2 and over L_2, it is convenient to separate the irreducible part of hyperpolarizability into two terms,

$$\gamma_j^{(\mathrm{non})}(nL_J)=\overline{\gamma}_j-\tilde{\gamma}_j. \tag{23}$$

After calculating the sums over total angular momenta of intermediate states, the expressions for the two components of the nonresonant hyperpolarizability of an atom in a triplet state may be written as follows:

$$\begin{aligned}\overline{\gamma}_j={}&24(-1)^{J+L+1}\Pi_L^2\Pi_{jJ}\left[\frac{(2J+1-j)_j}{(2J+2)_j}\right]^{1/2}\\ &\times\begin{Bmatrix}j & L & L\\ 1 & J & J\end{Bmatrix}\sum_{j_1j_2}\Pi_{j_1j_2}C^{j_10}_{10\,10}C^{j_20}_{10\,10}C^{j0}_{j_10\,j_20}\sum_{L_2}\Pi^2_{L_2}\\ &\times\begin{Bmatrix}L_2 & j_1 & L\\ j & L & j_2\end{Bmatrix}\sum_{L_1}C^{L_10}_{L0\,10}C^{L_10}_{L_20\,10}\begin{Bmatrix}L_1 & 1 & L_2\\ j_1 & L & 1\end{Bmatrix}\\ &\times\sum_{L_3}C^{L_30}_{L0\,10}C^{L_30}_{L_20\,10}\begin{Bmatrix}L_3 & 1 & L_2\\ j_2 & L & 1\end{Bmatrix}R^{1111}_{L_1L_2L_3}\end{aligned} \tag{24}$$

and

$$\begin{aligned}\tilde{\gamma}_j={}&24\Pi_L^4\Pi_J^2\Pi_{jJ}\left[\frac{(2J+1-j)_j}{(2J+2)_j}\right]^{1/2}\\ &\times\sum_{j_1j_2}\Pi_{j_1j_2}C^{j_10}_{10\,10}C^{j_20}_{10\,10}C^{j0}_{j_10\,j_20}\begin{Bmatrix}j_1 & j_2 & j\\ J & J & J\end{Bmatrix}\\ &\times\begin{Bmatrix}j_1 & L & L\\ 1 & J & J\end{Bmatrix}\times\begin{Bmatrix}j_2 & L & L\\ 1 & J & J\end{Bmatrix}\sum_{L_1}(C^{L_10}_{L0\,10})^2\\ &\times\begin{Bmatrix}L_1 & 1 & L\\ j_1 & L & 1\end{Bmatrix}R^{11}_{L_1}\sum_{L_3}(C^{L_30}_{L0\,10})^2\begin{Bmatrix}L_3 & 1 & L\\ j_2 & L & 1\end{Bmatrix}R^{101}_{L_3L_3}.\end{aligned} \tag{25}$$

Here the notation for the Nth-order radial matrix element is used,

$$R^{n_1n_2,\ldots,n_{N+1}}_{L_1L_2,\ldots,L_N}=\langle nL_J|r^{n_1}g_{L_1}r^{n_2}g_{L_2}r^{n_3}\cdots r^{n_N}g_{L_N}r^{n_{N+1}}|nL_J\rangle. \tag{26}$$

The Sturm-series resolution for the Green function in the FMP method (see the Appendix A) makes it possible to derive

TABLE IV. The numerical values for the components of polarizability $\alpha_{3\,^3P_J}$ and the nonresonant part of hyperpolarizability $\gamma_j(3\,^3P_J)^{(\mathrm{non})}$ of helium.

Component	Direct summation	Model potential
$\alpha^s_{3\,^3P_0}$ [4]	17203	17266
$\alpha^s_{3\,^3P_2}$ [4]	17193	17255
$\alpha^t_{3\,^3P_2}$ [4]	351.65	374.16
$\gamma_0^{(\mathrm{non})}(3\,^3P_0)$	-7.084×10^{11}	-7.077×10^{11}
$\gamma_0^{(\mathrm{non})}(3\,^3P_1)$	-7.073×10^{11}	-7.065×10^{11}
$\gamma_2^{(\mathrm{non})}(3\,^3P_1)$	-3.602×10^{10}	-3.573×10^{10}
$\gamma_0^{(\mathrm{non})}(3\,^3P_2)$	-7.069×10^{11}	-7.061×10^{11}
$\gamma_2^{(\mathrm{non})}(3\,^3P_2)$	7.142×10^{10}	7.079×10^{10}
$\gamma_4^{(\mathrm{non})}(3\,^3P_2)$	1.862×10^{8}	1.996×10^{8}

the hypergeometric-type series representation for the radial matrix element (26) which converges absolutely and may be calculated numerically to any desired precision. Typically, five to six terms in the series (A2) are sufficient for the relative accuracy of 10^{-5} [5,11]. The Sturm series account also for the contribution of the continuum, so a reliable control is ensured for the accuracy of calculations with the finite sums of oscillator strengths.

To verify the contribution of states neglected in the finite sums, we compared the numerical results for the radial matrix elements of second, third, and fourth order, which appear in calculations of hyperpolarizability components (24),(25), as computed in the FMP approximation with the use of the Green function and the finite sums of oscillator strengths without account for the continuum. The relative difference between the two values does not exceed 3×10^{-3} when both sums are computed to a relative accuracy not less than 10^{-4}. This is about the best possible precision for the sums over the discrete spectrum since, e.g., the triple summation is strictly divergent (the single and double summations are convergent) [16]. However, a few lowest terms in every sum usually form a convergent sequence which is sufficient to obtain the accuracy indicated above.

The numerical results for $\alpha_{3\,^3P_J}$ and $\gamma^{(\mathrm{non})}_{3\,^3P_J}$ are listed in Table IV. Only nonresonant contributions which may be used directly in Eq. (13) are presented. For calculating the resonant contributions, the data for the tensor polarizability and the expressions of Table III may be used. As seen from these data and Eq. (13), the relativistic CI method gives for the minimum of $\delta_{02}(F)$ the value $\overline{\delta_{02}}=8231$ MHz at the point of the $0^-\times0^-$ anticrossing at $\overline{F}=152.6$ kV/cm. The corresponding results given by the FMP approximation $\overline{\delta_{02}}=8234$ MHz and $\overline{F}=147.6$ kV/cm are in a good agreement with the preceding data. Finally, the field correction of the order of F^4 contributes about 2.7% of the total shift at the point of the anticrossing and gives a fairly large deviation from the quadratic Stark effect which may be essential within the limits of the experimental accuracy.

It should be noted that a calculation of the energy separation $\delta_{02}(F)$ by using Eq. (11), which includes the other excited states with $L\neq1$, leads to the result $\overline{\delta_{02}}=8244$ MHz. Independently, also by matrix diagonalization the result $\overline{\delta_{02}}=8249$ MHz was obtained earlier [3]. These theoretical data and the corresponding experimental value [3] $\overline{\delta_{02}}=8257\pm5$ MHz are not in a satisfying agreement. The difference between the theory and experiment is not acceptable and necessitates further investigations, both on the experimental and theoretical side.

As pointed out in the Introduction, the level of the experimental accuracy can be increased significantly by using more elaborate experimental techniques. On the other hand, the theoretical consideration of higher-order effects requires a development of the effective method of perturbation theory as applied to processes, taking into account both relativistic and correlation effects by an *ab initio* calculation of complicated spectral sums in the nonresonant case.

VI. CONCLUSIONS

In connection with laser spectroscopy measurements which have been carried out and are planned [3], we have calculated the higher-order dipole susceptibilities for the $1s3p\,^3P_0$ and $1s3p\,^3P_2$ states of helium. The usual assumption, that the major portion of the relativistic correction to the scalar polarizability is determined by the transition energy and not by the matrix elements, was also taken into account by combining precise variational calculations with the experimental transition energies. Simple expressions have been obtained which can be used to estimate the J dependence by calculations of the susceptibilities in the resonant approximation.

The present study may be important for further experimental investigations of the $0^-\times0^-$ anticrossing measurements in He I, where the experimental accuracy can be improved considerably, in particular, by using microwave spectroscopy. Another interesting example of the field-strength dependence, namely, the splitting $\delta_{12}(F)$ between the fine-structure sublevels with total angular momenta $J=1$ and $J=2$ and the magnetic number $M=1$ may also be studied on the basis of our data. On the other hand, the hyperpolarizabilities themselves are of current interest because of their relevance to optical harmonic generation through the interaction of intense beams of light with helium atoms.

ACKNOWLEDGMENTS

This work was supported by the Russian Foundation for Basic Research (RFBR Grant Nos. 96-02-00257 and 97-02-16407) and by the Deutsche Forschungsgemeinschaft [Grant No. 436 RUS 113/164/O(R,S)]. The work of A.D. and W.R.J. was supported in part by NSF Grant No. PHY 95-13179.

APPENDIX A: ANALYTICAL REPRESENTATION OF THE GREEN FUNCTION FOR THE FUES' MODEL POTENTIAL

The angular variables of the valence electron may be separated in the Green function $G_E(\vec{r}_1,\vec{r}_2)$ corresponding to the energy E by resolving into infinite series over a spherical-harmonic basis [5,11]:

$$G_E(\vec{r}_1,\vec{r}_2)=\sum_{lm} g_l(E;r_1,r_2)Y_{lm}(\vec{n}_1)Y^*_{lm}(\vec{n}_2), \tag{A1}$$

where $\vec{n}=\vec{r}/r$ is the unit vector, and the coefficients of these series are the radial Green functions, which in turn may be resolved into Sturm-function series,

$$g_l(E;r,r')=-\frac{4Z}{\nu}(xx')^l\exp\left(-\frac{x+x'}{2}\right)\times\sum_{k=0}^{\infty}\frac{k!L_k^{2\lambda_l+1}(x)L_k^{2\lambda_l+1}(x')}{\Gamma(k+2\lambda_l+2)(k+\lambda_l+1-\nu)}, \tag{A2}$$

where $x=2Zr/\nu$, $\nu=Z/(-2E)^{1/2}$, and $L_m^l(x)$ is the Laguerre polynomial. The poles of the radial Green function $g_l(E;r,r')$ correspond to the hydrogenlike energies $E_{nl}=-Z^2/2\nu_{nl}^2$ with $\nu=\nu_{nl}=n_r+\lambda_l+1$. Here, $n_r=0,1,2,\ldots$ is the radial quantum number and λ_l is the effective orbital momentum. The residues at these poles are equal to the products of corresponding radial eigenfunctions given by Eq. (19). Other representations for the Green function of the Fues model potential were given in [5].

Note that the series in Eq. (A2) converge for all r,r' (excluding the point $r=r'=0$) when $E<0$. The advantage of this representation is that it is similar to the representation of eigenfunctions in which effective wave functions for intermediate states are polynomials times exponentials, similar to the bound-state wave functions, but with one and the same argument for all of them. Such a representation includes the sum over bound states and the integral over continuum.

The reduced Green function, corresponding to the energy of a bound level E_{nl}, is defined by a limiting procedure

$$G^R(E_{nl};\mathbf{r},\mathbf{r}')=\frac{\partial}{\partial E}[(E-E_{nl})G(E;\mathbf{r},\mathbf{r}')],\quad E\to E_{nl}. \tag{A3}$$

The radial parts of $G^R(E_{nl};\mathbf{r},\mathbf{r}')$ are presented in [5].

The expression in the right-hand side of Eq. (A1) is particularly convenient when the contribution to the energy shift or the amplitude of a radiation process under consideration is given by a finite number of the partial terms in this expansion, for example, because of the selection rules.

APPENDIX B: RADIAL MATRIX ELEMENTS

We here present the formulas for the irreducible parts of hyperpolarizability (8) written in terms of the radial matrix elements (26), as calculated according to Eqs. (23)–(25). The expressions given below for the irreducible parts of hyperpolarizability differ from those of Ref. [4], where the fine-structure effects were not taken into account:

$$\gamma_0^{(\mathrm{non})}(n\,^3P_0)=\frac{8}{9}\left(R_{010}^{1111}-\frac{1}{3}R_0^{11}R_{00}^{101}\right)+\frac{32}{45}\left[2R_{210}^{1111}-\frac{5}{6}(R_2^{11}R_{00}^{101}+R_0^{11}R_{22}^{101})\right]+\frac{272}{225}\left[R_{212}^{1111}-\frac{50}{51}R_2^{11}R_{22}^{101}\right]+\frac{32}{25}R_{232}^{1111}, \tag{B1}$$

$$\gamma_0^{(\mathrm{non})}(n\,^3P_1)=\frac{8}{9}\left(R_{010}^{1111}-\frac{1}{2}R_0^{11}R_{00}^{101}\right)+\frac{32}{45}\left[2R_{210}^{1111}-\frac{7}{8}(R_2^{11}R_{00}^{101}+R_0^{11}R_{22}^{101})\right]+\frac{272}{225}\left[R_{212}^{1111}-\frac{67}{68}R_2^{11}R_{22}^{101}\right]+\frac{32}{25}R_{232}^{1111}, \tag{B2}$$

$$\gamma_2^{(\mathrm{non})}(n^3P_1)=\frac{4}{9}\left(R_{010}^{1111}-\frac{1}{2}R_0^{11}R_{00}^{101}\right)+\frac{16}{45}\left[2R_{210}^{1111}-\frac{7}{8}(R_2^{11}R_{00}^{101}+R_0^{11}R_{22}^{101})\right]+\frac{28}{225}\left[R_{212}^{1111}-\frac{13}{14}R_2^{11}R_{22}^{101}\right]+\frac{16}{175}R_{232}^{1111}, \tag{B3}$$

$$\gamma_0^{(\mathrm{non})}(n\,^3P_2)=\frac{8}{9}\left(R_{010}^{1111}-\frac{17}{30}R_0^{11}R_{00}^{101}\right)+\frac{32}{45}\left[2R_{210}^{1111}-\frac{107}{120}(R_2^{11}R_{00}^{101}+R_0^{11}R_{22}^{101})\right]+\frac{272}{225}\left[R_{212}^{1111}-\frac{1007}{1020}R_2^{11}R_{22}^{101}\right]+\frac{32}{25}R_{232}^{1111}, \tag{B4}$$

$$\gamma_2^{(\mathrm{non})}(n\,^3P_2)=-\frac{8}{9}\left(R_{010}^{1111}-\frac{25}{42}R_0^{11}R_{00}^{101}\right)-\frac{32}{45}\left[2R_{210}^{1111}-\frac{151}{168}(R_2^{11}R_{00}^{101}+R_0^{11}R_{22}^{101})\right]-\frac{56}{225}\left[R_{212}^{1111}-\frac{277}{294}R_2^{11}R_{22}^{101}\right]-\frac{32}{175}R_{232}^{1111}, \tag{B5}$$

$$\gamma_4^{(\mathrm{non})}(n\,^3P_2)=-\frac{8}{225\times 35}(5R_0^{11}+R_2^{11})(5R_{00}^{101}+R_{22}^{101}). \tag{B6}$$

[1] D. Kaiser, G. von Oppen, J. Phys. B **27**, 4835 (1994).
[2] O. Reusch, G. von Oppen, J. Phys. B **29**, 1681 (1996).
[3] R. Schumann, M. Dammasch, U. Eichmann, Y. Kriescher, G. Ritter, and G. von Oppen, J. Phys. B **30**, 2581 (1997).
[4] V. A. Davydkin and V. D. Ovsiannikov, J. Phys. B **17**, L207 (1984); **19**, 2071 (1986).
[5] N. L. Manakov, V. D. Ovsiannikov, and L. P. Rapoport, Phys. Rep. **141**, 319 (1986).
[6] W. R. Johnson, D. R. Plante, and J. Sapirstein, Adv. At., Mol., Opt. Phys. **35**, 255 (1995).
[7] M. H. Chen, K. T. Cheng, and W. R. Johnson, Phys. Rev. A **47**, 3692 (1993).
[8] K. T. Cheng, M. H. Chen, W. R. Johnson, and J. Sapirstein, Phys. Rev. A **50**, 247 (1994).
[9] A. Derevianko, Igor M. Savukov, W. R. Johnson, and D. R. Plante, Phys. Rev. A **58**, 4453 (1998).
[10] G. Simons, J. Chem. Phys. **55**, 756 (1971).
[11] N. L. Manakov, V. D. Ovsiannikov, and L. P. Rapoport, Opt. Spektrosk. **38**, 369 (1975) [Opt. Spectrosc. **38**, 206 (1975)].
[12] A. Derevianko, W. R. Johnson, V. D. Ovsiannikov, V. G. Pal'chikov, D. R. Plante, and G. von Oppen, Zh. Éksp. Teor. Fiz. **115**, 494 (1999) [Sov. Phys. JETP **88**, 272 (1999)].
[13] G. W. F. Drake, in *Handbook of Atomic, Molecular and Optical Physics*, edited by G. W. F. Drake (AIP Press, Woodbury, NY, 1996), Chap. 11.
[14] V. D. Ovsiannikov and S. V. Goossev, Phys. Scr. **57**, 506 (1998).
[15] D. A. Varshalovich, A. N. Moskalev, and V. K. Khersonskii, *Quantum Theory of Angular Momentum* (World Scientific, Singapore, 1988).
[16] N. L. Manakov, S. I. Marmo, and A. G. Fainshtein, Zh. Éksp. Teor. Fiz. **91**, 51 (1986) [Sov. Phys. JETP **64**, 29 (1986)].