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HYDROGEN ATOM IN CROSSED EXTERNAL FIELDS REEXEMINED BY THE MOMENT METHOD: Preprint ITEP 29-98/

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Recurrence relations of perturbation theory for hydrogen ground state are obtained. With their aid polarizabilities in constant perpendicular electric and magnetic fields are computed up to 80th order. The high orders asymptotic is compared with its quasiclassical estimate. For the case of arbitrary mutual orientation of external fields a general sixth order formula is given.

Fig.- 2, ref. - 20.

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### 1. Introduction

The hydrogen atom in constant homogeneous electric and magnetic fields still remains to be an object of theoretical investigations. A good example is the recent work [1], where a recurrent nonperturbative method is developed for building the exact wave function of hydrogen atom in magnetic field in the form of convergent double series. More wide discussion of the problem is contained in the review [2].

The famous technical trouble, namely the inability to separate the variables, only stimulate application of new investigation methods, including perturbative ones. The moment method [3], first used for perturbative treatment of the anharmonic oscillator, is not related with variables separation. The recent application of this method to the Zeeman effect problem [4] allowed to check the behavior of high orders asymptotic of the perturbation series. The moment method in the form similar to that used in [4], was developed independently of Ader's work [3] by Fernandez and Castro [5]. Then it was applied to hydrogen atom placed in parallel electric and magnetic fields [6] and later the Zeeman effect problem was considered for four sequences of hydrogen atom states [7].

It seems to be even more important to apply it to hydrogen atom in crossed electric  $(\vec{\mathcal{E}})$  and magnetic  $(\vec{\mathcal{H}})$  fields because only initial terms of expansion in powers of  $\vec{\mathcal{E}}$  and  $\vec{\mathcal{H}}$  were considered for this case up to now [8, 9, 10, 11, 12]. As will be shown here the moment method allows to compute high enough orders of this expansion.

The high orders asymptotic can be obtained with the help of the imaginary time method [13, 14, 15]. This asymptotic is determined by the contribution of an extreme subbarrier classical trajectory into the atom ionization probability [16, 17]. A pair of extreme paths replaces this trajectory at some value of the ratio of external fields  $\gamma = \mathcal{H}/\mathcal{E}$ . The dependence of far perturbation series terms on  $\gamma$  reflects this change of extreme trajectory and should be especially sharp for perpendicular external fields. We study here just this case.

#### 2. Recurrent evaluation of perturbation series

Consider the ground state of hydrogen atom, placed in perpendicular electric and magnetic fields. These fields are supposed to be constant and homogeneous. We restrict ourselves with nonrelativistic approximation and neglect the spin of electron. From the very beginning we take measures to simplify the numerical computations and to achieve high enough order of perturbation theory. For this aim we consider  $\gamma$  as a fixed parameter, replacing the double expansion in external fields by the single-variable series

$$\psi = \sum_{k=0}^{\infty} \mathcal{E}^k |k\rangle , \qquad E = \sum_{j=0}^{\infty} E_{2j}^{\perp} \mathcal{E}^{2j} , \qquad (1)$$

where the wave function corrections  $|k\rangle$  and hyper-polarizabilities  $E_k^{\perp}$  depend on  $\gamma$ . We introduce also circular coordinates

$$x_{\pm} = x \pm i y \; ,$$

then all further relations will have real coefficients. In these coordinates the hamiltonian of our problem is:

$$\hat{H} = \hat{H}_{0} + \mathcal{E}\hat{H}_{1} + \mathcal{E}^{2}\hat{H}_{2}; \qquad \hat{H}_{0} = -\frac{1}{2}\Delta - \frac{1}{r};$$

$$\hat{H}_{1} = x + \frac{\gamma}{2}\hat{L}_{z} = \frac{1}{2}(x_{+} + x_{-}) + \frac{\gamma}{2}\left(x_{+}\frac{\partial}{\partial x_{+}} - x_{-}\frac{\partial}{\partial x_{-}}\right); \qquad (2)$$

$$\hat{H}_{2} = \frac{\gamma^{2}}{8}(x^{2} + y^{2}) = \frac{\gamma^{2}}{8}x_{+}x_{-}.$$

The wave function correction of the order k satisfies the differential equation

$$(\hat{H}_0 - E_0)|k\rangle = -\hat{H}_1|k-1\rangle - \hat{H}_2|k-2\rangle + \sum_{j=1}^{[k/2]} E_{2j}^{\perp}|k-2j\rangle$$
 (3)

Just as in other problems where the moment method was used [3, 4] it is not difficult to transform equation (3) into algebraic relation between moments of the order k

$$P^{k}_{\sigma\alpha\beta} = \langle 0 | r^{\sigma-\alpha-\beta} x^{\alpha}_{+} x^{\beta}_{-} | k \rangle .$$
<sup>(4)</sup>

A recurrence relation results

$$\frac{1}{2}(\sigma - \alpha - \beta)(\sigma + \alpha + \beta + 1)P_{\sigma - 2,\alpha\beta}^{k} + 2\alpha\beta P_{\sigma - 2,\alpha - 1,\beta - 1}^{k} - \sigma P_{\sigma - 1,\alpha\beta}^{k} = R_{\sigma\alpha\beta}^{k}, \quad (5)$$

where

$$\begin{split} R^{k}_{\sigma\alpha\beta} &\equiv \frac{1}{2} [P^{k-1}_{\sigma+1,\alpha+1,\beta} + P^{k-1}_{\sigma+1,\alpha,\beta+1} + \gamma(\alpha-\beta)P^{k-1}_{\sigma\alpha\beta}] + \frac{\gamma^{2}}{8} P^{k-2}_{\sigma+2,\alpha+1,\beta+1} \\ &- \sum_{j=1}^{[k/2]} E^{\perp}_{2j} P^{k-2j}_{\sigma\alpha\beta} \; . \end{split}$$

The right-hand side of eq. (5) and hyper-polarizability  $E_k^{\perp}$  depend only on the moments of preceding orders. As usual in the moment method [3], the orthogonality condition is accepted

$$\langle 0|k\rangle = \delta_{0,k} \qquad \longrightarrow \qquad P_{0,0,0}^k = \delta_{0,k} . \tag{6}$$

An expression for hyper-polarizability arises from eq. (5) at  $\sigma = \alpha = \beta = 0$ and even k

$$E_k^{\perp} = \frac{1}{2} (P_{1,1,0}^{k-1} + P_{1,0,1}^{k-1}) + \frac{\gamma^2}{8} P_{2,1,1}^{k-2} .$$
<sup>(7)</sup>

The closed system of recurrence relations (5) - (7) allows to achieve, at least in principle, an arbitrary high order of perturbation theory. The sequence of operations is similar (also somewhat simpler) to that, used in the work [4] to compute Zeeman's shift of a non-degenerate state. At every order k only moments  $P_{\sigma\alpha\beta}^{k}$  from the sector  $\sigma \geq \alpha + \beta - 1$ ,  $\alpha \geq 0$ ,  $\beta \geq 0$ are necessary. They are evaluated by successively increasing of  $\sigma$ ,  $\alpha$  and  $\beta$ values with the help of eq. (5).

We have obtained hyper-polarizabilities in perpendicular fields up to 80th order, see table 1. This order is large enough to compare the dependence of these coefficients on  $\gamma$ , see fig. 1, with the predictions, following from quasiclassical considerations. One can see from fig. 1, that the function  $f_k(\gamma) \equiv \ln(|E_k^{\perp}|/k!)$  has two features. It has a minimum at  $\gamma \approx 3.4$  and a sequence of singular points to the right of this value. Besides, the function  $E_k^{\perp}(\gamma)$  changes it's sign at every singular point of  $f_k(\gamma)$ .

As follows from table 1, at not very large  $\gamma$  values all  $E_{2j}^{\perp}$  coefficients have negative sign, as in the case of Stark effect. In intermediate region of  $\gamma$  values the sequence of  $E_{2j}^{\perp}$  signs is irregular and for sufficiently large  $\gamma$ 's the series has normal Zeeman's sequence of signs  $(-1)^{j+1}$ .

## 3. High orders asymptotic

As is well known [16], a dispersion relation connects asymptotic of high orders coefficients  $E_k^{\perp}$  with the ionization probability of the atom i.e. with the penetrability of the potential barrier. This relation arises as a consequence of the fact, that the energy eigenvalue  $E = E_0(\mathcal{E}^2) - \frac{i}{2}\Gamma(\mathcal{E}^2)$  has essential singularity at  $\mathcal{E}^2 = 0$  and a cut along  $\mathcal{E}^2 > 0$  semiaxis. (And similarly  $E(\mathcal{H}^2)$  has essential singularity at  $\mathcal{H} = 0$  and a cut  $\mathcal{H}^2 < 0$ .)

To evaluate the ionization probability  $\Gamma$  the imaginary time method was previously developed [13, 14, 15]. The leading term of the asymptotic  $\tilde{E}_k^{\perp}$ of  $E_k^{\perp}$  coefficients at  $k \to \infty$  is determined by the classical subbarrier path with extremal value of the abbreviated action. Time takes complex values during this subbarier motion. There are two kinds of complex classical trajectories. Like in the Stark effect case, the ionization may be caused by electric field, at stabilizing influence of the magnetic field. The path of this kind creates the asymptotic

$$\tilde{E}_k^{\perp}(\gamma) \sim k! \, a^k(\gamma) \,, \quad k \text{ is even },$$
(8)

at not very large magnetic field, for  $\gamma$  below some critical value  $\gamma_c$ . According to [18]  $\gamma_c = 3.54$  for perpendicular external fields. And it is possible to cross the barrier also at  $\mathcal{H}^2 < 0$ , like in the Zeeman effect problem. Subbarrier trajectories of this kind are responsible for the form of  $\tilde{E}_k^{\perp}(\gamma)$  in the opposite case  $\gamma > \gamma_c$ . This change of asymptotic explains the origin of the left minimum in fig. 1.

Having in mind to get estimate for the function  $a(\gamma)$ , entering  $\vec{E}_k^{\perp}$ , we apply the results of [18, 19] and write here some necessary expressions for the special case of perpendicular external fields. More general considerations related to arbitrary  $\vec{\mathcal{E}}$  and  $\vec{\mathcal{H}}$  mutual orientation are contained in the work [18].

The time of subbarrier motion satisfies the equation [19]:

$$\tau^2 - (\tau c t h \tau - 1)^2 = \gamma^2 \tag{9}$$

which has a set of solutions  $\tau_n = in\pi + \tau'_n$ . The minimal value of the imaginary part of the subbarrier action is provided by  $\tau_0$  for  $\gamma < \gamma_c$  and

by a pair of solutions  $\tau_{\pm 1}$  for  $\gamma > \gamma_c$ . In the region  $\gamma < \gamma_c$  the energy half-width is

$$\Gamma(\mathcal{E}^2) = \frac{B(\gamma)}{\mathcal{E}} \exp\left[-\frac{2g(\gamma)}{3\mathcal{E}}\right], \quad g(\gamma) = \frac{3\tau}{2\gamma^3} \left(\gamma^2 - \sqrt{\tau^2 - \gamma^2}\right). \tag{10}$$

The dispersion relation in  $\mathcal{E}^2$  then leads to

$$\tilde{E}_{2j}^{\perp} = -\frac{1}{2\pi} \int_{0}^{\infty} \frac{\Gamma(z)dz}{z^{j+1}} \sim (2j)! \, a^{2j}, \tag{11}$$

where

$$a(\gamma) = \frac{3}{2g(\gamma)} . \tag{12}$$

The last equality is valid also in the region  $\gamma > \gamma_c$ , where  $g(\gamma)$  and  $a(\gamma)$  are complex functions. At  $\gamma < \gamma_c$  the resulting approximate expressions for  $a(\gamma)$  are

$$a(\gamma) = \frac{3}{2} \left( 1 - \frac{1}{30} \gamma^2 - \frac{71}{2100} \gamma^4 + \cdots \right) , \qquad \gamma \ll 1 ; \qquad (13)$$

$$a(\gamma) \simeq \frac{4\gamma^3}{(\gamma^2 - 1)^2(1 - 2e^{-\gamma^2 - 1})}, \qquad \gamma \gg 1.$$
 (14)

And in the region  $\gamma > \gamma_c$  another representation works

$$|a(\gamma)| = \frac{\gamma}{\pi} \left[ 1 - \frac{2}{\gamma^2} + \left( \frac{8\pi^2}{3} + 3 \right) \frac{1}{\gamma^4} + \cdots \right] \,. \tag{15}$$

On the other hand in the limit of large k a simple relation appropriate for numerical evaluation holds:

$$\ln|a(\gamma)| = \frac{d}{dk} \ln \frac{|E_k|}{k!} . \tag{16}$$

Evaluating  $a(\gamma)$  above  $\gamma_c$ , we used smoothed function  $E_k^{\perp}(\gamma)$ , with the nodes vicinities excluded. A comparison of this way numerically obtained function  $a(\gamma)$  with expressions (13) – (15) is presented in fig. 2.

Now we turn our attention to the region  $\gamma > \gamma_c$ . Two solutions of eq. (9)  $\tau_1$  and  $\tau_{-1}$  lead to complex conjugate values of  $g(\gamma)$ . Substituting approximate  $\tau_1$  value into second expression (10), it is possible to get the phase of the function  $a(\gamma)$ :

$$\arg\left(a\right)=-\arg\left(g\right)=-\frac{\pi}{2}+\alpha(\gamma),\qquad \alpha(\gamma)=\frac{2}{\gamma}-\frac{\pi^{2}+2}{3\gamma^{3}}+O(1/\gamma^{5})\;.$$

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Finally the sign-alternating asymptotic arises:

$$\tilde{E}_{2j}^{\perp} = 2 |B(\gamma)| (2j)! |a|^{2j+1} \cos\left[ (2j+1) \left( -\frac{\pi}{2} + \alpha(\gamma) \right) + \beta(\gamma) \right]$$
  
~  $(-1)^{j} (2j)! |a|^{2j+1} \sin\left[ (2j+1)\alpha(\gamma) + \beta(\gamma) \right], \quad j \gg 1.$  (17)

Here  $\beta(\gamma) = \arg(B)$  is the phase of the preexponential factor. Its relative contribution to the total phase falls like 1/j.

When the order of perturbation 2j is fixed and  $\gamma$  increases, expression (17) changes its sign at every point where the argument of the sinus turns to zero. This could explain the singular points in fig. 1 in the language of asymptotic. But rather lengthy calculations are required to establish detailed quantitative correspondence between asymptotic (17) and exact  $E_{2j}^{\perp}$  coefficients, including nodes vicinities. Simple approximate expression for  $\alpha(\gamma)$  is not enough for this aim.

### 4. Discussion

For the general case of the ground state energy expansion in powers of crossed external fields, the term of the forth power was known long enough [9].

$$E = -\frac{1}{2} + \sum_{j=1}^{\infty} E^{(2j)} ; \qquad E^{(2)} = -\frac{4}{9}\vec{\mathcal{E}}^2 + \frac{1}{4}\vec{\mathcal{H}}^2 ; \qquad (18)$$

$$E^{(4)} = -\frac{3555}{64}\vec{\mathcal{E}}^4 + \frac{159}{32}\vec{\mathcal{E}}^2\vec{\mathcal{H}}^2 + \frac{10}{3}[\vec{\mathcal{H}}\vec{\mathcal{E}}]^2 - \frac{53}{192}\vec{\mathcal{H}}^4 .$$
(19)

The value of  $E^{(4)}$  is confirmed for perpendicular fields by the work [12] and for parallel fields – by [12, 17, 20]. The  $E_4^{\perp}$  coefficient, computed by means of recurrence relations (5) – (7) exactly agree with (19). But we have noticed numerical difference between our coefficient  $E_6^{\perp}$  and corresponding quantity from the work [12]. Therefore the sixth order of perturbation theory was analyzed in details.

The magneto-electric susceptibilities, i.e. coefficients of the double series in powers of external fields, can be easily obtained from hyper-polarizabilities  $E_k^{\perp}(\gamma)$ . Thus, in the sixth order, taking into account that Stark's and Zeeman's coefficients are fixed, it is enough to choose

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four different  $\gamma$  values and to solve the system of four linear equations. The following representation results

$$E_{6}^{\perp} = -\frac{1}{512} \left( 2512779 - 521353\gamma^{2} + \frac{953869}{27}\gamma^{4} - \frac{5581}{9}\gamma^{6} \right)$$
$$\equiv \sum_{j=0}^{3} \gamma_{6-2j,2j}^{\perp} (\mathcal{H}/\mathcal{E})^{2j} .$$
(20)

(The last identity introduces notation of [12].) Using linear relation between expansions (1) and (18) and the known magneto-electric susceptibilities in parallel fields [20], it is easy to obtain another term of series (18):

$$E^{(6)} = -\frac{2512779}{512}\vec{\mathcal{E}}^6 + \frac{254955}{512}\vec{\mathcal{E}}^4\vec{\mathcal{H}}^2 + \frac{133199}{256}\vec{\mathcal{E}}^2[\vec{\mathcal{H}}\vec{\mathcal{E}}]^2 -\frac{49195}{1536}\vec{\mathcal{E}}^2\vec{\mathcal{H}}^4 - \frac{255557}{6912}\vec{\mathcal{H}}^2[\vec{\mathcal{H}}\vec{\mathcal{E}}]^2 + \frac{5581}{4608}\vec{\mathcal{H}}^6 .$$
(21)

Some next terms of series (18) can be obtained in the same way. Expressions (20) and (21) are convenient to check term by term the sixth order correction. As follows from [12]

$$\gamma_{24}^{\perp[12]} = -\frac{1610197}{27648} \text{ and } \gamma_{42}^{\perp[12]} = \frac{2417015}{1536} ,$$
 (22)

while the results of our computation are

$$\gamma_{24}^{\perp} = -\frac{953869}{13824} \quad \text{and} \quad \gamma_{42}^{\perp} = \frac{521353}{512} .$$
 (23)

All other corresponding coefficients of [12] and of present work coincide. We carried out additional independent calculation by means of the method from the work [9] and get

$$\gamma_{24}^{\perp[9]} = -\frac{953869}{13824} , \qquad (24)$$

see Appendix. Note, that [9] contains complete correction of the sixth power in external fields for the case of parallel fields and only a part of it for the case of perpendicular fields. These "celebrated" sixth order terms result as a by-product of forth-order calculations in the work [9]. The agreement between high-order hyper-polarizabilities  $E_k^{\perp}$  and their asymptotic  $\tilde{E}_k^{\perp}$  presents additional confirmation of correctness of recurrence relations (5) – (7).

### 5. Concluding remarks

The considered above problem demonstrates once more the high efficiency and convenience of the moment method. The obtained recurrence relations have allowed to advance up to 80th order of perturbation theory. Besides the unusual "oscillations" of hyper-polarizabilities as a function of the ratio of external fields were noticed. The high orders asymptotical behavior was analyzed as well. Basic parameters of this asymptotic exactly agree with those, previously obtained on the ground of quasiclassical approximation with the help of imaginary time method.

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# Appendix

Extending the described in the work [9] calculations we obtained, by the same method, the ground state energy correction, which is proportional to  $Q \equiv \mathcal{H}^2[\mathcal{H}\vec{\mathcal{E}}]^2$ . The perturbation in [9] includes Stark's term  $V_s = \vec{\mathcal{E}}\vec{r}$ , paramagnetic  $V_p = \frac{1}{2}\mathcal{H}\vec{\mathcal{L}}$  and diamagnetic  $V_D = \frac{1}{8}[\mathcal{H}\vec{r}]^2$  terms. The entire perturbation is inhomogeneous, therefore terms of the sixth power in external fields are presented in corrections of the fourth, fifth and sixth orders in V.

$$\varepsilon^{(4)} = -\frac{151347}{2047}Q + \dots,$$
(A1)

$$\varepsilon^{(5)} = \langle 2|(V - \varepsilon^{(1)})|2\rangle - 2\varepsilon^{(2)}\langle 2|1\rangle - \varepsilon^{(3)}\langle 1|1\rangle , \qquad (A2)$$

$$\varepsilon^{(6)} = \langle 3|(V - \varepsilon^{(1)})|2\rangle - \varepsilon^{(2)}(\langle 2|2\rangle + \langle 1|3\rangle) - 2\varepsilon^{(3)}\langle 2|1\rangle - \varepsilon^{(4)}\langle 1|1\rangle .$$
 (A3)

In the following an abbreviated mnemonic notation will be used, reflecting the origin of each term and the powers of entering this term external fields. This notation helps to omit all not essential terms. Operation signs are encircled in the abbreviated notation. In the first order in V

$$|1\rangle \equiv \{a_1(r)(\vec{\mathcal{E}}\vec{r}) + \frac{1}{4}(a_2(r)\mathcal{H}^2 + a_3(r)[\vec{\mathcal{H}}\vec{\mathcal{E}}]^2)\}|0\rangle \bigoplus \{V_{\mathcal{E}} \bigoplus V_D\}|0\rangle . \quad (A4)$$

The next correction  $|2\rangle$  contains

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$$V_P V_{\mathcal{E}}|0\rangle \bigoplus_{i=1}^{i} b_1(r)([\vec{\mathcal{H}}\vec{\mathcal{E}}]\vec{r})|0\rangle ,$$
$$V_{\mathcal{E}} V_D|0\rangle \bigoplus_{i=1}^{i} \{(\vec{\mathcal{E}}\vec{r})(b_4(r)[\vec{\mathcal{H}}\vec{r}]^2 + b_5(r)\mathcal{H}^2) + b_6(r)\mathcal{H}^2([\vec{\mathcal{H}}\vec{\mathcal{E}}]\vec{r})\}|0\rangle .$$
(A5)

The polynomials  $a_i(r)$  and  $b_i(r)$  are given in the article [9]. Abbreviated notation allows to verify that in each of the right-hand sides of Eqs. (A2) and (A3) only the first matrix element yields contribution  $\sim Q$ .

$$\varepsilon^{(5)} = A + B_1 + B_2 + C + \dots$$

dots stand for all omitted terms.

$$A \bigoplus \langle 0 | (V_P V_{\mathcal{E}}) V_D (V_P V_{\mathcal{E}}) | 0 \rangle ,$$
  

$$B_1 \bigoplus \langle 0 | (V_{\mathcal{E}} V_D) V_P (V_P V_{\mathcal{E}}) | 0 \rangle ,$$
  

$$B_2 \bigoplus \langle 0 | (V_P V_{\mathcal{E}}) V_P (V_{\mathcal{E}} V_D) | 0 \rangle , \quad B_2 = B_1 ,$$
  

$$C \bigoplus -\varepsilon^{(1)} \langle 0 | (V_P V_{\mathcal{E}}) (V_P V_{\mathcal{E}}) | 0 \rangle .$$
(A6)

Corresponding explicit expressions are

$$\begin{split} A &= \frac{1}{32} \langle 0 | b_1^2(r) ([\vec{\mathcal{H}}\vec{\mathcal{E}}]\vec{r})^2 [\vec{\mathcal{H}}\vec{r}]^2 | 0 \rangle = \frac{571}{48} Q , \\ B_1 &= \frac{1}{16} \langle 0 | b_1(r) \{ b_4(r) [\vec{\mathcal{H}}\vec{r}]^2 + (b_5(r) + b_6(r)) \mathcal{H}^2 \} ([\vec{\mathcal{H}}\vec{\mathcal{E}}]\vec{r})^2 | 0 \rangle \\ &= \frac{299623}{18432} Q = B_2 , \\ C &= -\frac{1}{16} \mathcal{H}^2 \langle 0 | b_1^2(r) ([\vec{\mathcal{H}}\vec{\mathcal{E}}]\vec{r})^2 | 0 \rangle = -\frac{9673}{4608} Q . \end{split}$$
(A7)

Only one term of the third correction to the wave function is essential – that of the lowest power in external fields:

$$|3\rangle = \frac{1}{4}c_1(r)([\vec{\mathcal{H}}[\vec{\mathcal{H}}\vec{\mathcal{E}}]]\vec{r})|0\rangle + ... \bigoplus (V_P V_{\mathcal{E}} \bigoplus ...)|0\rangle .$$
(A8)

The differential equation for  $c_1(r)$  [9] is satisfied by the polynomial:

$$c_1(r) = -\frac{1}{144} (450 + 225r + 62r^2 + 6r^3) . \tag{A9}$$

As a consequence we get

$$\varepsilon^{(6)} \bigoplus \langle 0 | (V_P^2 V_{\mathcal{E}}) V_P (V_P V_{\mathcal{E}}) | 0 \rangle + \dots$$
 (A10)

 $\mathbf{and}$ 

$$\varepsilon^{(6)} = -\frac{1}{16} \langle 0|b_1(r)c_1(r)([\vec{\mathcal{H}}[\vec{\mathcal{H}}\vec{\mathcal{E}}]]\vec{r})^2|0\rangle + \dots .$$
 (A10')

The total energy correction of the desired form is

$$\Delta E = -\frac{255557}{6912} \mathcal{H}^2 [\vec{\mathcal{H}}\vec{\mathcal{E}}]^2 + \dots .$$
 (A11)

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One should not forget also the "isotropic" contribution to the energy correction, originating from  $\varepsilon^{(4)}$ :

$$-\frac{49195}{1536}\mathcal{H}^4\mathcal{E}^2$$
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k	$\gamma = 2.0$	$\gamma = 3.0$	$\gamma = 6.0$	$\gamma = 70.0$
2	-1.2500000	+0.0000000	+6.7500000	+1222.7500
4	-26.755208	-3.1875000	-114.42188	-6587135.8
6	-1861.2023	-449.50781	-1167.7324	$+1.4083939 \times 10^{11}$
8	-231011.83	-39518.994	+3563855.9	$-5.5211341  imes 10^{15}$
10	$-4.3046334 \times 10^{7}$	-4415104.3	$-1.9148046 \times 10^{9}$	$+3.2420587  imes 10^{20}$
12	$-1.1108858 \times 10^{10}$	$-7.8928562 \times 10^{8}$	$+8.7798001 \times 10^{11}$	$-2.6154136  imes 10^{25}$
14	$-3.7903062 \times 10^{12}$	$-1.9681752  imes 10^{11}$	$-2.7563534  imes 10^{14}$	$+2.7647695 \times 10^{30}$
16	$-1.6565997 \times 10^{15}$	$-6.0102169 \times 10^{13}$	$-1.4386041 \times 10^{17}$	$-3.7128281  imes 10^{35}$
18	$-9.0515867 \times 10^{17}$	$-2.2599584  imes 10^{16}$	$+5.1094372 \times 10^{20}$	$+6.1877660 \times 10^{40}$
20	$-6.0598915 \times 10^{20}$	$-1.0569584 \times 10^{19}$	$-8.5724488 \times 10^{23}$	$-1.2555439  imes 10^{46}$
22	$-4.8865029 \times 10^{23}$	$-5.9768835  imes 10^{21}$	$+1.0371317 \times 10^{27}$	$+3.0513954 \times 10^{51}$
24	$-4.6763388  imes 10^{26}$	$-3.9866393  imes 10^{24}$	$-3.8968989 \times 10^{29}$	$-8.7572953 \times 10^{56}$
26	$-5.2434742 \times 10^{29}$	$-3.1103268 \times 10^{27}$	$-3.2330568 \times 10^{33}$	$+2.9313099 \times 10^{62}$
28	$-6.8121442  imes 10^{32}$	$-2.8159706 \times 10^{30}$	$+1.5576520 \times 10^{37}$	$-1.1320123  imes 10^{68}$
30	$-1.0154266 \times 10^{36}$	$-2.9246470 \times 10^{33}$	$-4.7085047 \times 10^{40}$	$+4.9954972 \times 10^{73}$
40	$-4.4829424  imes 10^{52}$	$-2.1028668 \times 10^{49}$	$-6.2371218  imes 10^{58}$	$-4.7347888 \times 10^{102}$
50	$-2.3374671 \times 10^{70}$	$-1.7764719 \times 10^{66}$	$-8.3813757 \times 10^{77}$	$+4.7994225 \times 10^{132}$
60	$-8.8335861 \times 10^{88}$	$-1.0843471 \times 10^{84}$	$-5.8480016 \times 10^{97}$	$-3.1030447 \times 10^{163}$
70	$-1.7441216  imes 10^{108}$	$-3.4513657 \times 10^{102}$	$+1.9127476 \times 10^{117}$	$+8.3761299 \times 10^{194}$
80	$-1.4229765 \times 10^{128}$	$-4.5336207 \times 10^{121}$	$+1.3366049 \times 10^{140}$	$-4.3789967  imes 10^{226}$

Table 1. Hyper-polarizabilities  $E_k^{\perp}$  of the hydrogen ground state.



Fig. 1. Functions  $f_k(\gamma) = \ln \frac{|E_k^{\perp}|}{k!}$  resulting from the recurrently computed hyper-polarizabilities.





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