Strongly forbidden two-photon transitions in atoms

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The strongly forbidden two-photon transition $1S_{1/2} \to 2P_{1/2}$ between hyperfine components with total angular momentum $F = 0$ is considered. Estimates are obtained of the parity-nonconservation effects produced in this transition by the interaction between the electrons and the nucleus. The results are generalized to include multielectron atoms.

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1. INTRODUCTION

Allowed two-photon transitions in atoms are being intensively investigated at present.[1] It can be hoped that progress in laser techniques will permit investigation also of forbidden two-photon transitions, such as $1S_{1/2} \to 2P_{1/2}$ in hydrogen. The nonrelativistic amplitude of the $1S_{1/2} \to 2P_{1/2}$ two-photon transition in the hydrogen atom was obtained in Refs. 2 and 3. In this approximation, the amplitude of the two-photon transition between the hyperfine components of the $1S_{1/2}$ and $2P_{1/2}$ levels with total angular momentum $F = 0$ (the amplitude of the transition $0^+ \to 0^-$) vanishes. A nonzero result is obtained when account is taken of the relativistic corrections to this amplitude.

The amplitude of the transition $1S_{1/2}\ (F=0) \to 2P_{1/2}(F=0)$ contributes to the absorption of two photons of equal frequency; the contribution is investigated experimentally by the method of Dopplerless spectroscopy. The amplitude is also of interest for the study of parity-nonconservation effects that are connected with weak interactions of the electrons with the atomic nucleus.[3]

In Secs. 2 and 3 of this paper we calculate the amplitude of the two-photon transition $1S_{1/2}(F=0) \to 2P_{1/2}(F=0)$ in the first nonvanishing approximation, using a relativistic calculation. The solution for the hydrogen atom is obtained in analytic form.

The two-photon transition amplitude is generalized in Sec. 4 to include multielectron atoms with one nS electron in the outer shell. In Sec. 5 we obtain a quantitative estimate of the parity-nonconservation effect upon absorption of two photons of like helicity and equal frequency from colliding beams.

2. AMPLITUDE OF THE TRANSITION $1S_{1/2}$ $(F=0) \to 2P_{1/2}$ $(F=0)$

We denote by A_{12} the amplitude of the two-photon transition between the states $|\psi_1\rangle$ and $|\psi_2\rangle$. The probability dw_{12} of the Raman scattering and the probability W_{12} of absorption of two photons are connected with the amplitude A_{12} by the relations

$$dw_{12}=J_1|A_{12}|^2\frac{d^3k_2}{(2\pi)^3}2\pi\delta(E_2-E_1+\omega_2-\omega_1),$$
$$W_{12}=J_1J_2|A_{12}|^2 2\gamma_{12}[(E_2-E_1-\omega_1-\omega_2)^2+\gamma_{12}^2]^{-1}. \quad (1)$$

Here J_1 and J_2 are the photon flux densities in the beams, γ_{12} is the arithmetic mean of the widths of the initial and final states, and ω_1 and ω_2 are the photon energies ($\hbar = c = 1$).

We calculate the matrix element (ME) M connected with the amplitude A_{12} by the relation

$$A_{12}=4\pi\alpha(4\omega_1\omega_2)^{-1/2}M, \quad \alpha=1/137,$$
$$M=\langle\psi_2|\tilde{A}_2G\tilde{A}_1|\psi_1\rangle+\langle\psi_2|\tilde{A}_1G\tilde{A}_2|\psi_1\rangle, \quad (2)$$
$$\tilde{A}=\alpha\mathbf{A}, \quad \mathbf{A}=\mathbf{e}e^{i\mathbf{kr}}, \quad \mathbf{ek}=0.$$

Here $\mathbf{A}$ is the vector potential of the photon, α are Dirac matrices, and G is the Green's function of the electron in the atomic field. The ME (2) is written for absorption of two photons. The scattering case is obtained from (2) by replacing $\tilde{A}_2$ by $\tilde{A}_2^*$, which is equivalent to making in the final result the substitutions $\mathbf{e}_2, \mathbf{k}_2, \omega_2 \to \mathbf{e}_2^*, -\mathbf{k}_2, -\omega_2$.

We consider first the two-photon transition $1S_{1/2}(F=0) \to 2P_{1/2}(F=0)$ in a hydrogenlike atom (F is the total angular momentum of the atom). In this case

$$\langle\psi_2|Q|\psi_1\rangle=\frac{1}{2}\sum_{m_1}\langle 2P_{1/2}, m_1|Q|1S_{1/2}, m_1\rangle, \quad (3)$$

where $|S_{1/2}, m_1\rangle$ and $|2P_{1/2}, m_1\rangle$ are the wave functions with the hyperfine structure nglected, m_1 is the projection of the electron angular momentum j, and Q stands for $\tilde{A}_2G\tilde{A}_1$ or $\tilde{A}_1G\tilde{A}_2$.

We consider a region of photon energies of the order of the ground-state ionization energy. In this case $\mathbf{k}\cdot\mathbf{r} \sim \omega r \sim \alpha Z \ll 1$, and the exponentials in (2) can be expanded in powers of $\mathbf{k}\cdot\mathbf{r}$. It was shown in Ref. 3 that the amplitude of this transition has a smallness $\sim(\alpha Z)^3$ compared with the amplitude of the allowed transition, so that the expansion of each exponential in (2) should extend to terms $\sim(\mathbf{k}\cdot\mathbf{r})^3$. For transitions between states with opposite negative parity, the ME containing even powers of $(\mathbf{k}\cdot\mathbf{r})$ vanish. As a result we get

$$M=M_1+M_2+M_3,$$
$$M_1=i\langle\psi_2|\tilde{e}_2\mathbf{k}_2\mathbf{r}G\tilde{e}_1+\tilde{e}_2G\tilde{e}_1\mathbf{k}_1\mathbf{r}|\psi_1\rangle+(1\leftrightarrow 2),$$
$$M_2=-\frac{i}{2}\langle\psi_2|\tilde{e}_2(\mathbf{k}_2\mathbf{r})^2G\tilde{e}_1\mathbf{k}_1\mathbf{r}+\tilde{e}_2(\mathbf{k}_2\mathbf{r})G\tilde{e}_1(\mathbf{k}_1\mathbf{r})^2|\psi_1\rangle+(1\leftrightarrow 2), \quad (4)$$
$$M_3=-\frac{i}{6}\langle\psi_2|\tilde{e}_2(\mathbf{k}_2\mathbf{r})^3G\tilde{e}_1+\tilde{e}_2G\tilde{e}_1(\mathbf{k}_1\mathbf{r})^3|\psi_1\rangle+(1\leftrightarrow 2),$$

where $(1 \leftrightarrow 2)$ denotes the interchange $(\mathbf{e}_1, \mathbf{k}_1, \omega_1) \rightleftarrows \mathbf{e}_2, \mathbf{k}_2, \omega_2)$.

The main difficulty lies in the calculation of the ME linear in $\mathbf{k}\cdot\mathbf{r}$ (M_1). The main contributions from these ME ($\sim\alpha Z$) are cancelled out by an analogous contribution from the cross terms, marked by the symbol $(1 \leftrightarrow 2)$. M_1 must therefore be calculated with account

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taken of the relativistic corrections, so as to preserve the terms $\sim(\alpha Z)^3$. The ME M_2 and M_3 already have the required order $(\alpha Z)^3$ and can be calculated in the nonrelativistic approximation.

a. Calculation of ME linear in k, r (M_1)

To calculate the ME contained in M_1 we need the relativistic Coulomb Green's function. We can use for this purpose the expression obtained in Refs. 4 and 5. As will be shown below, however, it is possible to transform M_1 in such a way that the required accuracy will be reached using a nonrelativistic Green's function. We use for this purpose the following expression obtained in the Appendix for the ME M_1:

$$M_1=\omega_1\omega_2\left\{\frac{\mathbf{s}_2}{E+E_2}\langle\psi_2|\sigma+\mathbf{L}-V[\alpha\mathbf{r}]|\Phi_1\rangle\right.$$
$$\left.+\frac{\mathbf{s}_1}{E+E_1}\langle\Phi_2|\sigma+\mathbf{L}-V[\alpha\mathbf{r}]|\psi_1\rangle\right\}+(1\leftrightarrow 2),$$
$$\mathbf{s}_i=[\mathbf{e}_i\mathbf{v}_i],\quad \mathbf{v}_i=\mathbf{k}_i/\omega_i\quad (i=1,2). \tag{5}$$

Here E_1, E_2, and E are the energies of the electron in the initial, final, and intermediate states,

$$E_2=E_1+\omega_1+\omega_2,\quad E=E_1+\omega_1=E_2-\omega_2, \tag{6}$$

V is the potential energy of the electron. We have introduced in (5) the auxiliary state vectors

$$|\Phi_1\rangle=G\mathbf{e}_1\mathbf{r}|\psi_1\rangle,\quad \langle\Phi_2|=\langle\psi_2|\mathbf{e}_2\mathbf{r}G. \tag{7}$$

The state vectors (7) can be written in the coordinate representation in the form of sums over spherical spinors

$$\langle\mathbf{r}|\Phi_1\rangle=\frac{1}{r}\sum_{jlm}a_{jl}^{mm_1}(S_\varkappa-iR_\varkappa\tilde{n})\begin{pmatrix}\Omega_{jlm}\\0\end{pmatrix}, \tag{8}$$

$$\langle\Phi_2|\mathbf{r}\rangle=\frac{1}{r}\sum_{jlm}b_{jl}^{m_1m}(\Omega^+_{jlm},0)(P_\varkappa+iL_\varkappa\tilde{n}), \tag{9}$$
$$\tilde{n}=\alpha\mathbf{n},\quad \mathbf{n}=\mathbf{r}/r,$$

where the radial functions are solutions of the equations

$$D_r\begin{pmatrix}S_\varkappa\\R_\varkappa\end{pmatrix}=-r\begin{pmatrix}f_S\\g_S\end{pmatrix},\quad D_r\begin{pmatrix}P_\varkappa\\L_\varkappa\end{pmatrix}=-r\begin{pmatrix}f_{1/2}\\g_{1/2}\end{pmatrix}, \tag{10}$$

$$D_r=\begin{pmatrix}\frac{d}{dr}+\frac{\varkappa}{r}, & V-E-m\\ V-E+m, & -\frac{d}{dr}+\frac{\varkappa}{r}\end{pmatrix},\quad \varkappa=\begin{cases}l & \text{if } j=l-1/2\\ -l-1 & \text{if } j=l+1/2\end{cases}. \tag{11}$$

Here $g_S(g_{1/2})$ and $f_S(f_{1/3})$ are the large and small components of the radial state function $1S_{1/2}$ $(2P_{1/2})$:

$$\langle\mathbf{r}|\psi_1\rangle=\frac{1}{r}(g_S-if_S\tilde{n})\begin{pmatrix}\Omega_{1/2m_1}\\0\end{pmatrix},$$
$$\langle\psi_2|\mathbf{r}\rangle=\frac{1}{r}(\Omega^+_{1/2m_1},0)(g_{1/2}+if_{1/2}\tilde{n}). \tag{12}$$

The coefficients a and b in (8) and (9) are given by the formulas

$$a_{jl}^{mm_1}=\int d\mathbf{n}\Omega^+_{jlm}\mathbf{e}_1\mathbf{n}\Omega_{1/2m_1},$$
$$b_{jl}^{m_1m}=\int d\mathbf{n}\Omega^+_{1/2m_1}\mathbf{e}_2\mathbf{n}\Omega_{jlm}, \tag{13}$$

with the $a_{jl}^{mm_1}$ different from zero only at $l=1, j=\frac{1}{2}$ and $\frac{1}{3}$, while $b_{jl}^{m_1m}$ differ from zero at $l=0, j=\frac{1}{2}$ and $l=2, j=\frac{3}{2}$.

The solutions of Eqs. (10) can be written in the form of the series

$$\begin{pmatrix}S_\varkappa\\R_\varkappa\end{pmatrix}=\sum_n\frac{I_{n\varkappa}}{E-E_{n\varkappa}}\begin{pmatrix}g_{n\varkappa}\\f_{n\varkappa}\end{pmatrix}\qquad \begin{pmatrix}P_\varkappa\\L_\varkappa\end{pmatrix}=\sum_n\frac{I_{n\varkappa}}{E-E_{n\varkappa}}\begin{pmatrix}g_{n\varkappa}\\f_{n\varkappa}\end{pmatrix} \tag{14}$$

where

$$J_{n\varkappa}=\int_0^\infty dr(g_{n\varkappa}g_S+f_{n\varkappa}f_S),$$
$$I_{n\varkappa}=\int_0^\infty dr(g_{1/2}g_{n\varkappa}+f_{1/2}f_{n\varkappa}), \tag{15}$$

where $g_{n\varkappa}$ and $f_{n\varkappa}$ are the large and small components of the radial state function with quantum numbers n, j, and l.

Using the expansions (8), (9), and (14) and the orthogonality of the radial wave functions, we transform the ME (5) into

$$M_1=-\frac{2}{9}\omega_1\omega_2\left\{\frac{\mathbf{e}_1\mathbf{s}_2}{E+E_2}\left[-\frac{J_{1/2}}{E-E_{1/2}}+\frac{1}{2}\frac{J_{1/2}}{E-E_2}\right.\right.$$
$$+\int_0^\infty dr[\Delta gS_{-2}+f_{1/2}R_1+f_{1/2}R_{-2}+rVg_{1/2}(R_1-R_{-2})$$
$$\left.+rVf_{1/2}(S_1-S_{-2})]\right]+\frac{\mathbf{e}_2\mathbf{s}_1}{E+E_1}\left[\frac{3}{2}\frac{J_{1/2}}{E-E_1}\right.$$
$$\left.\left.+\int_0^\infty dr[(L_2-L_1)(f_S+rVg_S)+(P_2-P_{-1})rVf_S]\right]\right\}+(1\leftrightarrow 2), \tag{16}$$

where

$$\Delta g=g_{1/2}+g_{3/2},\quad J_j=\int_0^\infty dr(g_jg_S+f_jf_S)\quad (j=1/2,3/2), \tag{17}$$

$g_{3/2}$ and $f_{3/2}$ are the components of the wave function of the $2P_{3/2}$ state, and $E_{3/2}$ is the energy of this state.

We consider first the terms of (16) outside the integral and designate them by M_1'. In our order in αZ, the energies of the states $2P_{3/2}$ and $2P_{1/2}$ are equal to $E_{3/2}\approx E_2$, and their wave functions differ in sign, $g_{3/2}\approx -g_{1/2}$, so that $J_{3/2}\approx -J_{1/2}$. Therefore the principal terms $\sim\alpha Z$ in M_1' cancel out the like cross terms, and only terms of order $(\alpha Z)^3$ are left as a result:

$$M_1'=-\frac{2}{9}\omega_1\omega_2\left\{\frac{\mathbf{e}_1\mathbf{s}_2}{E+E_2}\left[-\frac{J_{1/2}}{E-E_{1/2}}+\frac{1}{2}\frac{J_{1/2}}{E-E_2}\right]\right.$$
$$\left.+\frac{3}{2}\frac{\mathbf{e}_2\mathbf{s}_1}{E+E_1}\frac{J_{1/2}}{E-E_1}\right\}+(1\leftrightarrow 2)\approx\frac{\omega_1\omega_2}{6m^2}J\left\{\mathbf{e}_1\mathbf{s}_2\right.$$
$$\left.\times\left[\frac{I^2}{\omega_2}+\frac{1}{2}-\frac{2}{3}\frac{m}{\omega_2}\left(\frac{\Delta E}{\omega_2}+\frac{\Delta J}{J}\right)\right]-\mathbf{e}_2\mathbf{s}_1\left(\frac{I_1}{\omega_1}-\frac{1}{2}\right)\right\}+(1\leftrightarrow 2). \tag{18}$$

Here $I_1=m-E_1$; $I_2=m-E_2$ are the ionization energies of the initial and final states; $\Delta E=E_{3/2}-E_{1/2}$ is the fine splitting of the levels $2P_{1/2}$ and $2P_{3/2}$; $\Delta J=J_{1/2}+J_{3/2}$; J is the nonrelativistic limit of the integral $J_{1/2}$; g_P is the nonrelativistic limit of $g_{1/2}$,

$$J=\int_0^\infty drrg_Pg_S. \tag{19}$$

The integral terms in (16) are already of the required order of smallness. It suffices to express in them the small components of the functions in terms of the large ones and take the latter to equal their nonrelativistic values

$$\begin{pmatrix} f_{n\varkappa} \\ R_\varkappa \\ L_\varkappa \end{pmatrix} \approx \frac{1}{2m}\left(\frac{d}{dr}+\frac{\varkappa}{r}\right)\begin{pmatrix} g_{n\varkappa} \\ S_\varkappa \\ P_\varkappa \end{pmatrix}. \tag{20}$$

The functions $S_\varkappa$ and $P_\varkappa$ in (20) satisfy the inhomogeneous Schrödinger equation

$$\left(\frac{d^2}{dr^2}-\frac{\varkappa(\varkappa+1)}{r^2}+p^2-2mV\right)\begin{pmatrix} S_\varkappa \\ P_\varkappa \end{pmatrix}=2mr\begin{pmatrix} g_S \\ g_P \end{pmatrix},$$
$$p^2=E^2-m^2\approx -2m(I_1-\omega_1)\approx -2m(I_2+\omega_2). \tag{21}$$

It is seen from (21) that in the relativistic limit $S_l=S_{-l-1}$ and $P_L=P_{-l-1}$. With the aid of (18) and (20) we rewrite the ME (16) in the form

$$M_1=\frac{\omega_1\omega_2}{6m^2}\left\{\mathbf{e}_2\mathbf{s}_1\left(\frac{1}{2}-\frac{I_1}{\omega_1}\right)J+\mathbf{e}_1\mathbf{s}_2\left[\frac{1}{2}+\frac{I_2}{\omega_2}\right.\right.$$
$$\left.-\frac{2}{3}\frac{m}{\omega_2}\left(\frac{\Delta E}{\omega_2}+\frac{\Delta J}{J}\right)\right]J-\frac{1}{3}\mathbf{e}_2\mathbf{s}_1\int_0^\infty dr\left[\frac{3}{r}P_2 j_s\right.$$
$$+\left(4V+r\frac{dV}{dr}+I_1\right)P_0 g_S-\left(V+r\frac{dV}{dr}+I_1\right)P_2 g_S\Big]$$
$$\left.-\frac{1}{3}\mathbf{e}_1\mathbf{s}_2\int_0^\infty dr\left[2m\Delta g+3Vg_P\right]S_1\right\}+(1\leftrightarrow 2). \tag{22}$$

Formula (22) is suitable for $0^+\to 0^-$ transitions in any central field. In the case of a Coulomb field $V=-\alpha Z/r$ formula (22) can be simplified to

$$M_1=\frac{\omega_1\omega_2}{6m^2}\left\{\mathbf{e}_2\mathbf{s}_1\left(\frac{1}{2}-\frac{4}{3}\frac{I_1}{\omega_1}\right)J+\mathbf{e}_1\mathbf{s}_2\left[\frac{1}{2}+\frac{I_2}{\omega_2}\right.\right.$$
$$\left.-\frac{2}{3}\frac{m}{\omega_2}\left(\frac{\Delta E}{\omega_2}+\frac{\Delta J}{J}\right)\right]J+\mathbf{e}_2\mathbf{s}_1 I_1\int_0^\infty dr\left(\frac{1}{3}P_2+\frac{P_2+2P_0}{\eta r}\right)g_S$$
$$\left.+\mathbf{e}_1\mathbf{s}_2\int_0^\infty dr\left(\frac{\alpha Z}{r}g_P-\frac{2}{3}m\Delta g\right)S_1\right\}+(1\leftrightarrow 2), \tag{23}$$
$$\eta=m\alpha Z.$$

In the derivation of (23) we took into account the equality $\int_0^\infty dr P_0 g_S=J/\omega_1$.

An important factor in (22) and (23) is that they contain only the nonrelativistic functions $S_\varkappa$ and $P_\varkappa$, which are expressed in terms of the nonrelativistic Green's function in an external field. The relativistic values are needed only for the wave functions of the states $1S_{1/2}$, $2P_{1/2}$, and $2P_{3/2}$ in the calculation of ΔJ and Δg.

b. Calculation of the ME cubic in k · r (M_2 and M_3)

The expressions (4) from which the ME M_2 and M_3 are calculated already contain the required smallness $\sim(\alpha Z)^3$, so that the transition to the nonrelativistic approximation is effected in M_2 and M_3 directly. The Green's function G in M_2 and M_3 contains a sum over both the positive and the negative frequencies.

In the nonrelativistic approximation the sum over the negative frequencies is calculated by replacing G by $(1-\beta)/4m$. It can be shown that for the $0^+\to 0^-$ transition the negative ME parts $M_2^{(-)}$ and $M_3^{(-)}$ are equal to zero independently of the character of the transition.

In the calculation of that part of the ME which corresponds to the sum over positive frequencies, the transition to the nonrelativistic approximation is via the substitution

$$\langle\psi_n|\hat{A}|\psi_k\rangle\to\frac{1}{m}\langle\varphi_n|\mathbf{Ap}+\frac{1}{2}\boldsymbol{\sigma}\,\mathrm{rot}\,\mathbf{A}|\varphi_k\rangle, \tag{24}$$

where $\mathbf{p}=-i\nabla$; φ_n and φ_k are the nonrelativistic values of the functions ψ_n and ψ_k.

In the calculation of M_2 and M_3 we shall need also the nonrelativistic values of the functions Φ_1 and Φ_2 [Eqs. (8) and (9)]

$$\langle\mathbf{r}|\Phi_1^{\mathrm{nr}}\rangle=\frac{1}{r}\mathbf{e}_1\mathbf{n}S_1\chi_{m_1}(4\pi)^{-1/2}, \tag{25}$$

$$\langle\Phi_2^{\mathrm{nr}}|\mathbf{r}\rangle=-(4\pi)^{-1/2}\chi_{m_1}^+\frac{1}{r}\left\{\frac{1}{3}\boldsymbol{\sigma}\mathbf{e}_2(P_0-P_2)+(\boldsymbol{\sigma}\mathbf{n})(\mathbf{e}_2\mathbf{n})P_2\right\}, \tag{26}$$

where χ_{m_1} are Pauli spinors and $\mathbf{n}=\mathbf{r}/r$.

Using the transformations (A.3)–(A.5) and changing to the nonrelativistic limit in accord with (24) we obtain, taking (25) and (26) into account, the following expressions for M_2 and M_3:

$$M_2=-\mathbf{e}_2\mathbf{s}_1\frac{\omega_2^2}{12m^2}J+\frac{\omega_1^2\omega_2^2}{60m}\left\{[2(\mathbf{e}_2\mathbf{s}_1)(\mathbf{v}_2\mathbf{v}_1)\right.$$
$$+\mathbf{e}_1\mathbf{s}_2]\int_0^\infty dr r^2 g_P S_1+[2(\mathbf{e}_1\mathbf{s}_2)(\mathbf{v}_1\mathbf{v}_2)$$
$$\left.+\mathbf{e}_2\mathbf{s}_1]\int_0^\infty dr r^2 P_2 g_S\right\}+(1\leftrightarrow 2), \tag{27}$$

$$M_3=\frac{\omega_1\omega_2}{12m}\left\{\frac{1}{3}(\mathbf{e}_2\mathbf{s}_1)\omega_1^2\int_0^\infty dr r^2\left(P_0-\frac{2}{5}P_2\right)g_S\right.$$
$$\left.+\frac{1}{5}(\mathbf{e}_1\mathbf{s}_2)\omega_2^2\int_0^\infty dr r^2 g_P S_1\right\}+(1\leftrightarrow 2). \tag{28}$$

In the calculation of M_2 there appear in place of the functions Φ_1 and Φ_2 the functions

$$|\Phi_1'\rangle=G^{np}\mathbf{k}_1\mathbf{r}|\varphi_1\rangle,\quad \langle\Phi_2'|=\langle\varphi_2|\mathbf{k}_2\mathbf{r}G^{np}. \tag{29}$$

Their values are obtained from (25) and (26) by the substitution $\mathbf{e}_i\to\mathbf{k}_i$, as can be readily verified by comparing (29) with (7).

Combining the formulas (22), (27), and (28) we obtain the sought matrix element M (4). It contains radial integrals of the nonrelativistic wave functions and of the functions S and P that satisfy the Schrödinger equation (21). For an arbitrary field, these integrals can be calculated by numerical methods. In the case of the hydrogen atom the problem can be solved exactly and can be carried through to conclusion analytically.

3. HYDROGENLIKE ATOMS

All the calculations in this section are made for the Coulomb field $V=-\alpha Z/r$. With the aid of (23), (27), and (28) we express the matrix element M in the form

$$M=\mathbf{e}_2\mathbf{s}_1 T(\omega_1,\omega_2)+(\mathbf{e}_2\mathbf{s}_1)(\mathbf{v}_2\mathbf{v}_1)Q(\omega_1,\omega_2)+(1\leftrightarrow 2),$$
$$\mathbf{v}_i=\mathbf{k}_i/\omega_i,\quad \mathbf{s}_i=[\mathbf{e}_i\mathbf{v}_i]. \tag{30}$$

The functions $T(\omega_1,\omega_2)$ and $Q(\omega_1,\omega_2)$ are given by

$$T(\omega_1,\omega_2)=\frac{\omega_1\omega_2}{6m^2}\left\{\left[1-\frac{12}{12}\frac{I_1}{\omega_1}-\frac{\omega_2}{2\omega_1}-\frac{2m}{3\omega_1}\left(\frac{\Delta J}{J}+\frac{\Delta E}{\omega_1}\right)\right]J\right.$$
$$+\alpha Z\left(J_1+\frac{1}{2}J_3+\frac{1}{6}\eta J_4+J_6'\right)-\frac{2}{3}mJ_8'$$
$$\left.+\frac{m\omega_1^2}{6}\left[J_2-J_5+\frac{9}{20}\frac{I_1}{\omega_1}(J_5+J_7')\right]\right\}, \tag{31}$$

$$Q(\omega_1,\omega_2)=\frac{\omega_1^2\omega_2^2}{30m}(J_5'+J_7), \tag{32}$$

where

$$J_1=\langle P_0|r^{-1}|g_S\rangle,\quad J_4=\langle P_2|g_S\rangle,\quad J_7=\langle g_P|r^2|S_1\rangle,$$
$$J_2=\langle P_0|r^2|g_S\rangle,\quad J_5=\langle P_2|r^2|g_S\rangle,\quad J_8=\langle \Delta g|S_1\rangle,$$
$$J_3=\langle P_2|r^{-1}|g_S\rangle,\quad J_6=\langle g_P|r^{-1}|S_1\rangle,\quad J=\langle g_P|r|g_S\rangle,$$
$$\Delta J=\langle \Delta g|r|g_S\rangle+\langle \Delta f|r|f_S\rangle,$$
$$I_1=m\alpha^2Z^2/2,\quad \Delta E=m\alpha^4Z^4/32,\quad \eta=m\alpha Z. \tag{33}$$

In formulas (33) we have used the notation

$$\langle\psi|r^n|\Phi\rangle=\int_0^\infty dr r^n\psi(r)\Phi(r). \tag{34}$$

The integrals J_i' are obtained from J_i by making the change $\omega_1\to\omega_2$.

The simplest matrix elements J and ΔJ are calculated by direct substitution of the corresponding wave functions

$$\Delta J=\frac{1}{4}\alpha^2Z^2\left(\ln\frac{3}{2}-\frac{1}{3}\right)J,\quad J=-\frac{12}{\eta}\left(\frac{2}{3}\right)^{n/2}. \tag{35}$$

To calculate the remaining matrix elements J_1–J_9 we need the explicit forms of the functions S_l and P_l, which are the solutions of the inhomogeneous Schrödinger equation (21). We use the nonrelativistic Green's function $G_l(r,r')$, which satisfies the equation

$$\left(\frac{d^2}{dr^2}-\frac{l(l+1)}{r^2}-\frac{2\eta}{r}+p^2\right)G_l(r,r')=\delta(r-r'),\quad p^2=E^2-m^2 \tag{36}$$

and is of the form[4,5]

$$G_l(r,r')=-(rr')^{1/2}\int_0^\infty\left(\operatorname{th}\frac{x}{2}\right)^{-2\nu}\exp\{-q(r+r')\operatorname{ch}x\}$$
$$\times I_{2l+1}(2q(rr')^{1/2}\operatorname{sh}x)\,dx,$$
$$q=-ip,\quad \nu=i\alpha ZE/p=\eta/q,\quad \eta=m\alpha Z, \tag{37}$$

I_{2l+1} is a modified Bessel function of the first kind.

The solutions of (21) are expressed in terms G_l in accord with the formula

$$\begin{pmatrix}S_l(r)\\P_l(r)\end{pmatrix}=2m\int_0^\infty dr'G_l(r,r')r'\begin{pmatrix}g_S(r')\\g_P(r')\end{pmatrix}. \tag{38}$$

After substituting (37) and (38) in (33), the matrix elements J_1–J_8 are transformed into triple integrals. The double integration with respect to r and r' is carried out by means of the usual formulas (e.g., Eqs. 6.631.1 and 7.621.5 of Ref. 6). The remaining single integrals, after making the change of variable $\tanh(x/2)=t^{1/2}$ and after straightforward but cumbersome transformations can be expressed in terms of three simple integrals F, F_1, and F_2:

$$F=(3\nu)^3(\nu^2-1)\int_0^1\frac{t^{2-\nu}dt}{(a-bt)^3},$$

$$F_i=(3\nu)^4(\nu^2-1)\int_0^1\frac{t^{2-\nu}dt}{(a-bt)^3(a_i+b_it)}\quad(i=1,2),$$

where $a_1=1+\nu$, $a_2=1+\nu/2$, $a=a_1a_2$, $b_1=1-\nu$, $b_2=1-\nu/2$, $b=b_1b_2$, and $\nu=\eta/q=(1-\omega_1/I_1)^{-1/2}$. The final expressions for T and Q are:

$$T(\omega_1,\omega_2)=\frac{(\alpha Z)^3}{4m}\left(\frac{2}{3}\right)^{n/2}\left\{\frac{1}{8\Omega_1}-\frac{5}{12}-\frac{11}{4}\Omega_1\right.$$
$$\left.+2\Omega_1^2+F_1'+F_2+\frac{1}{4}\left(\frac{1}{6}+5\Omega_1\right)F'-\frac{3}{2}F\right\},$$
$$Q(\omega_1,\omega_2)=\frac{(\alpha Z)^3}{4m}\left(\frac{2}{3}\right)^{n/2}\Omega_1\{2-2\Omega_1-F-4F'\},\quad \Omega_i=\omega_i/I_1,\quad \Omega_1+\Omega_2=\tfrac{3}{4} \tag{39}$$

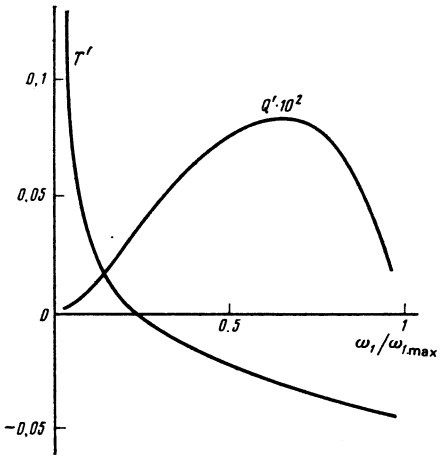

FIG. 1. Dependence of the amplitudes of $T'=m(\alpha Z)^{-3}T$ and $Q'=m(\alpha Z)^{-3}Q$ on the photon frequency ω_1.

The photon energies in (39) are in units of the ground-state ionization potential I_1. The integrals F' and F_i' are obtained from F and F_i by the substitution $\omega_1\to\omega_2$. The dependences of the amplitudes of T and Q on Ω_1 are shown in Fig. 1.

If the energy of one of the photons is low, e.g., $\Omega_2\ll\Omega_1$, but still $\Omega_2\gg\Delta E/I_1=\alpha^2Z^2/16$, then the matrix element M (2) takes the form

$$M\approx\frac{(\alpha Z)^3}{32m}\left(\frac{2}{3}\right)^{n/2}\left\{e_1s_2\left(\frac{1}{\Omega_2}+4\ln\frac{3}{2}-\frac{19}{6}\right)-\frac{27}{2}e_2s_1+O(\Omega_2)\right\}. \tag{40}$$

This formula can be obtained both from (39) and directly from (2) by summing over the intermediate states. When $\Omega_2\to0$, a nonzero contribution to the transition amplitude is made only by the intermediate states $1S_{1/2}$, $2S_{1/2}$, $2P_{1/2}$, and $2P_{3/2}$. The term $\sim\Omega_2^{-1}$ in (40) stems from the expansion of the energy denominator in the intermediate state $2P_{3/2}$.

4. MULTIELECTRON ATOMS

The results obtained in Sec. 2 can be applied to multielectron atoms (atoms of alkali metals Cu, Ag, and Au, and ions of certain elements) which have, one nS electron on top of the closed shell. Only a few of these atoms have terms with total atom angular momentum $F=0$. Examples of such atoms are the stable silver isotopes ^{107}Ag and ^{109}Ag, and certain ions.[7] For these, the formulas of Sec. 2 describe the two-photon transition $nS_{1/2}(F=0)\to n'P_{1/2}(F=0)$ at arbitrary frequencies and momenta of the photons. However, if we are interested only in absorption of two photons of equal frequency from colliding beams, the results can be applied to transitions between the levels $nS_{1/2}$ and $n'P_{1/2}$ with arbitrary but equal angular momenta F. Let us explain the foregoing.

In the most general case (without expansion in $\mathbf{k}\cdot\mathbf{r}$) the amplitude of the two-photon transition $nS_{1/2}\to n'P_{1/2}$ contains eight scalar functions T_i and can be written in the form

$$A(S_{1/2}\to P_{1/2})=i\frac{4\pi\alpha}{(4\omega_1\omega_2)^{1/2}}\frac{\alpha Z}{m}\{(\mathbf{e}_2\mathbf{e}_1)(\boldsymbol{\sigma}\mathbf{v}_1)T_1$$
$$+(\mathbf{e}_2\mathbf{v}_1)(\boldsymbol{\sigma}\mathbf{e}_1)T_2+ie_2s_1T_3+(\mathbf{e}_2\mathbf{v}_1)(\mathbf{e}_1\mathbf{v}_2)(\boldsymbol{\sigma}\mathbf{v}_1)T_4+(1\leftrightarrow2)\},$$
$$\mathbf{v}_i=\mathbf{k}_i/\omega_i,\quad \mathbf{s}_i=[\mathbf{e}_i\times\mathbf{v}_i],\quad T_i=T_i(\omega_1,\omega_2,\mathbf{v}_1\mathbf{v}_2), \tag{41}$$

where $\sigma/2$ is the electron angular momentum of the transition. The amplitudes of the transitions between the hyperfine components of the levels are obtained by projecting (41) on the corresponding hyperfine states. For transitions between levels $S_{1/2}$ and $P_{1/2}$ with $F=0$, all the terms of (41) that contain the electron angular momentum of the transition drop out, and only the term with T_3, which was calculated in Sec. 2, is left. As seen from (41), the same takes place when photons of equal frequency ($\omega_1=\omega_2$) from colliding beams ($\nu_1=-\nu_2$) are absorbed, independently of the total angular momentum F of the initial and final states. Interest attaches here only to transitions without change of F, for otherwise the entire transition amplitude vanishes. The photons in the beams should be circularly polarized and have an identical helicity, so as to exclude absorption of two photons from one beam.

In the case of atoms with large nuclear charge Z, the expression obtained in Sec. 2 for the matrix element M (4) can be greatly simplified. For an optical electron going from the ground to an excited state, the nuclear charge is strongly screened and the important role in the matrix elements of the transition is played by $\mathbf{k}\cdot\mathbf{r}\sim\alpha$. Therefore the integral terms in (23), (27), and (28) are of the order of α^3, just as in the hydrogen atom. At the same time the fine splitting $\Delta E=E(P_{3/2})-E(P_{1/2})$ of the P levels increases rapidly with increasing nuclear charge Z (in hydrogen, for example, $\Delta E\approx 0.36\ \text{cm}^{-1}$ and in cesium $\Delta E\approx 554\ \text{cm}^{-1}$, Ref. 7). The reason is that the energy of the spin-orbit interaction responsible for the fine splitting is proportional to $\langle r^{-1}dV/dr\rangle\sim\langle r^{-3}\rangle$ and is therefore determined by the region of short distances, where the nuclear charge is weakly screened. As a result, for heavy atoms the terms linear in $\mathbf{k}\cdot\mathbf{r}$ outside the integral are not cancelled out as in the case when the P levels are degenerate, and make the principal contribution to the matrix element M.

Retaining in (16) only the terms outside the integral, we obtain a simple approximate expression for the matrix element of the two-photon transition $nS_{1/2}(F=0)\to n'P_{1/2}(F=0)$ in a heavy atom:

$$M\approx -\mathbf{e}_1\mathbf{s}_2\frac{\omega_1}{9m}\left(J_{1/2}+\frac{\omega_2}{\omega_2+\Delta E}J_{3/2}\right)+(1\leftrightarrow 2), \tag{42}$$

$$J_j=\langle n'P_j|r|nS_{1/2}\rangle,\quad \Delta E=E(n'P_{3/2})-E(n'P_{1/2}). \tag{43}$$

When photons of like helicity and frequency are absorbed from colliding beam, formula (42), as explained above, is suitable for $nS_{1/2}\to n'P_{1/2}$ transitions between hyperfine levels with arbitrary equal angular momenta at the start and at the end. Only the absorption of E_1 and M_1 photons is taken into account in (42).

5. EFFECTS OF PARITY NONCONSERVATION IN TWO-PHOTON TRANSITIONS

The weak interaction of the electrons with the nucleus deprives the atomic levels of a definite parity. Interference of amplitudes with different parity lead to a dependence of the probability of the atomic transition on such pseudoscalar quantities as the helicity (the sign of the circular polarization) of the photon. It was shown in Ref. 3 that large parity-nonconservation effects can be expected for the two-photon transition $1S_{1/2}\to 2P_{1/2}$ in the hydrogen atom, if photons of like parity and helicity are absorbed from colliding laser beams. The parity nonconservation in such a transition manifests itself in a dependence of the absorption on the circular polarization of the photons. Order-of-magnitude estimates[3] show that the relative difference between the probabilities of the absorption of right- and left-polarized photons in this transition can reach $\sim 10^{-4}$–10^{-5}. The results obtained in Sec. 5 make possible an exact calculation of this value.

According to Ref. 3, for transitions between the $1S_{1/2}$ and $2P_{1/2}$ levels of hydrogen with total angular momentum F we have

$$\Delta_F\equiv\frac{W_R-W_L}{W_R+W_L}=-\delta_F\frac{T_0}{\alpha T_3}, \tag{44}$$

where δ_F is the value of mixing of the $2P_{1/2}$ and $2S_{1/2}$ states with angular momentum F:

$$\delta_0=-1.4\cdot 10^{-11}(\varkappa_1-3\varkappa_2),\quad \delta_1=-1.2\cdot 10^{-11}(\varkappa_1+\varkappa_2), \tag{45}$$

$\varkappa_1$ and $\varkappa_2$ are the weak constants of the electron-proton interaction. In the Weinberg–Salam theory, at an experimental value $\sin^2\theta_W\approx 0.22$ we have $\varkappa_1=\frac{1}{2}(1-4\sin^2\theta_W)\approx 0.06$ and $\varkappa_2=-1.25\varkappa_1\approx -0.075$, therefore

$$\delta_0\approx -4\cdot 10^{-12},\quad \delta_1\approx 1.8\cdot 10^{-13}.$$

The quantity T_0 in (44) is connected with the amplitude of the admixture two-photon transition $1S_{1/2}\to 2S_{1/2}$, while αT_3 is connected with the amplitude of the transition $1S_{1/2}\to 2P_{1/2}$. In the notation of the present paper we have at $\omega_1=\omega_2=(3/16)m\alpha^2$, taking (39) into account,

$$\begin{aligned} T_0&=2\sqrt{3}\left(\frac{2}{3}\right)^{11/2}(5F+1),\\ \alpha T_3&=m(T-Q)=\frac{\alpha^3}{4}\left(\frac{2}{3}\right)^{11/2}\left\{F_1+F_2+\frac{85}{96}F-\frac{125}{96}\right\}\end{aligned} \tag{46}$$

where F, F_1, and F_2 are defined in (38) at $\Omega=\frac{3}{8}$

$$F=0.0966,\quad F_1=0.0456,\quad F_2=0.0515,$$

from which we get

$$T_0/\alpha T_3=-4.7\cdot 10^7.$$

As a result,

$$\Delta_0\approx -1.9\cdot 10^{-4},\quad \Delta_1\approx 0.8\cdot 10^{-5}.$$

For the analogous transitions in deuterium ($F=\frac{1}{2},\frac{3}{2}$), the parity nonconservation effects are enhanced by the additional weak interaction of the electron with the neutron of the nucleus. Calculations yield

$$\Delta_{1/2}\approx -2.6\cdot 10^{-4},\quad \Delta_{3/2}\approx -2.7\cdot 10^{-4}.$$

Using the results of Sec. 4 we can obtain analogous parity-nonconservation effects in heavy atoms. To this end, however, it is necessary to know (e.g., from experiment) the amplitudes of the allowed two-photon transitions that become mixed-in with the ground transition on account of the weak interactions.

In conclusion the authors than R. M. Ryndin for helpful discussions of the questions touched upon in the paper.

APPENDIX

To transofrm the matrix elements that enter in M_1 (4) we use the operator equations

$$(H-E)G=-1, \quad H=\alpha p+\beta m+V, \qquad H|\psi_1\rangle=E_1|\psi_1\rangle, \quad \langle\psi_2|H=E_2\langle\psi_2| \tag{A.1}$$

and the identities

$$\alpha=i(Hr-rH), \qquad \tilde{e}\mathbf{kr}=\frac{i}{2}\{H(\mathbf{er})(\mathbf{kr})-(\mathbf{er})(\mathbf{kr})H\}+\frac{1}{2}[\mathbf{e}\times\mathbf{k}][\alpha\times\mathbf{r}], \tag{A.2}$$

$$H[\alpha\times\mathbf{r}]+[\alpha\times\mathbf{r}]H=-2(\sigma+\mathbf{L})+2V[\alpha\times\mathbf{r}].$$

With their aid one can prove the following equalities:

$$G\tilde{e}|\psi_1\rangle=i\{(E-E_1)G-1\}\mathbf{er}|\psi_1\rangle, \qquad \langle\psi_2|\tilde{e}G=i\langle\psi_2|\mathbf{er}\{1-(E-E_2)G\}, \tag{A.3}$$

$$G\tilde{e}\mathbf{kr}|\psi_1\rangle=\frac{i}{2}\{(E-E_1)G-1\} \times(\mathbf{er})(\mathbf{kr})|\psi_1\rangle+\frac{1}{2}[\mathbf{ek}]G[\alpha\mathbf{r}]|\psi_1\rangle,$$

$$\langle\psi_2|\tilde{e}\mathbf{kr}G=\frac{i}{2}\langle\psi_2|(\mathbf{er})(\mathbf{kr})\{1-(E-E_2)G\} +\frac{1}{2}[\mathbf{ek}]\langle\psi_2|[\alpha\mathbf{r}]G, \tag{A.4}$$

$$G[\alpha\mathbf{r}]|\psi_1\rangle=\frac{2}{E+E_1}\left\{-G(\sigma+\mathbf{L})+GV[\alpha\mathbf{r}]+\frac{1}{2}[\alpha\mathbf{r}]\right\}|\psi_1\rangle,$$

$$\langle\psi_2|[\alpha\mathbf{r}]G=\frac{2}{E+E_2}\langle\psi_2|\left\{-(\sigma+\mathbf{L})G+[\alpha\mathbf{r}]VG+\frac{1}{2}[\alpha\mathbf{r}]\right\}. \tag{A.5}$$

Here $E_2=E_1+\omega_1+\omega_2$ and $E=E_1+\omega_1=E_2-\omega_2$.

The purpose of the transformations (A.3)–(A.5) is to introduce, by changing the power of r, the required smallness directly into the expression from which the matrix element is calculated. The expression for M_1 (4) can be rewritten with the aid of (A.3) in the form

$$M_1=-\omega_2\langle\psi_2|\mathbf{e}_2\mathbf{r}G\tilde{e}_1(\mathbf{k}_1\mathbf{r})|\psi_1\rangle -\omega_1\langle\psi_2|\tilde{e}_2(\mathbf{k}_2\mathbf{r})G\mathbf{e}_1\mathbf{r}|\psi_1\rangle+(1\leftrightarrow 2). \tag{A.6}$$

The terms that did not contain G were cancelled out in (A.6) by the cross terms. We apply the transformation (A.4) to (A.6) and take into account the fact that the following matrix elements vanish for the $0^+\to 0^-$ transition

$$\langle\psi_2|(\mathbf{e}_2\mathbf{r})(\mathbf{e}_1\mathbf{r})(\mathbf{k}_1\mathbf{r})|\psi_1\rangle=0, \quad \langle\psi_2|\mathbf{e}_2\mathbf{r}G(\mathbf{e}_1\mathbf{r})(\mathbf{k}_1\mathbf{r})|\psi_1\rangle=0, \tag{A.7}$$

$$\langle\psi_2|(\mathbf{e}_2\mathbf{r})(\mathbf{k}_2\mathbf{r})(\mathbf{e}_1\mathbf{r})|\psi_1\rangle=0, \quad \langle\psi_2|(\mathbf{e}_2\mathbf{r})(\mathbf{k}_2\mathbf{r})G\mathbf{e}_1\mathbf{r}|\psi_1\rangle=0. \tag{A.8}$$

Indeed, in the $0^+\to 0^-$ transition the matrix elements (A.7) are pseudoscalars made up of vectors $\mathbf{e}_2$, $\mathbf{e}_1$, and $\mathbf{k}_1$. However, the only nonzero pseudoscalar $\mathbf{e}_2\cdot\mathbf{e}_1\times\mathbf{k}_1$ is antisymmetric in $\mathbf{e}_1$ and $\mathbf{k}_1$, in contrast to the matrix element (A.7). The vanishing of the matrix element (A.8) can be proved similarly. As a result we get

$$M_1=-\frac{\omega_1\omega_2}{2}\{\mathbf{s}_1\langle\psi_2|(\mathbf{e}_2\mathbf{r})G[\alpha\mathbf{r}]|\psi_1\rangle +\mathbf{s}_2\langle\psi_2|[\alpha\mathbf{r}]G(\mathbf{e}_1\mathbf{r})|\psi_1\rangle+(1\leftrightarrow 2)\}, \tag{A.9}$$

$$\mathbf{s}_i=[\mathbf{e}_i\mathbf{v}_i], \quad \mathbf{v}_i=\mathbf{k}_i/\omega_i.$$

Applying now the transformation (A.5) and recognizing that the following matrix elements vanish after integration over the angles

$$\langle\psi_2|[\alpha\mathbf{r}](\mathbf{e}_i\mathbf{r})|\psi_1\rangle=0 \quad (i=1,2), \tag{A.10}$$

we obtain expression (5) for M_1.

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