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TWO-PHOTON BOUND-BOUND TRANSITIONS IN A COULOMB FIELD

B. A. ZON, N. L. MANAKOV and L. I. RAPOPORT

Voronezh State University

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A method is proposed, which can be employed for obtaining analytic expressions of two-photon process amplitudes in a hydrogen atom, involving bound-bound transitions. The method is based on the use of a special integral representation of the Coulomb Green's function. The amplitude of the two-photon process can be expressed in terms of a hypergeometric function. As examples, cross sections are given for coherent scattering of light from the 1s, 2s and 2p levels and for Raman scattering involving the 1s ⇄ 2s and 1s ⇄ 3d transitions.

1. INTRODUCTION

RECENTLY, in connection with the development of laser technology, more interest is evinced in multiphoton processes occurring when a laser beam interacts with atoms. The solution of multiphoton problems for an arbitrary atom is a very complicated matter, and it is therefore of interest to trace the features of such processes using the simplest atom, hydrogen, and use the result as a basic model for the solution of problems involving more complicated atoms. Multiphoton processes in hydrogen itself are also of independent interest (particularly in problems of theoretical astrophysics). However, even in the case of the hydrogen atom, for which simple analytic expressions for the eigenfunctions and eigenvalues are available, we are faced with the need for summation of matrix elements of the electromagnetic-interaction operator over a complete set of intermediate states, including the continuous spectrum, and this entails certain difficulties.

Two-photon processes on hydrogen were considered in many papers. Thus, in[1,2] they calculated the probability of two-photon decay of the metastable 2s level by using an approximate numerical series summation. An approximate formula (semiquantitative) was obtained in[3] for the coherent scattering of light by a hydrogen atom in the ground state. The Schwartz-Tiemann me method[4] was used in[5] to find the cross section of the same process for incident-photon frequencies $0 \leq \hbar\omega \leq \frac{3}{4}$Ry. The Schwartz-Tiemann method was used also in[6] to calculate the Raman scattering of light for the 2s → 1 transition, and in[7] for the two-photon ionization of the 2s level for a limited frequency interval.

In our preceding paper[8], which was devoted to two-photon decay of the 2s level, we proposed a method that yields exact analytic expressions for the probabilities of two-photon processes in the Coulomb field for the nonrelativistic case.

In the present paper we use this method to derive a general expression for the probability of two-photon bound-bound transitions. An essential part of the method is the use of a convenient integral representation for the radial part of the Green's function of a charged particle in the Coulomb field. The analytic expressions derived in the paper for the transition amplitudes make it possible, by using crossing symmetry and the analyticity of the S matrix with respect to energy, to obtain the amplitude of one of the processes, and to calculate the amplitude of any other process for the same initial and final states.

In Sec. 2 we describe the method of calculating the two-photon transition amplitudes. By way of examples of the use of the general formula, we present in Sec. 3 an expression for the probability of coherent scattering light from the levels 1s, 2s, and 2p and of Raman scattering of light with the atomic transitions 1s → 2s and 1s → 3d. A concise content of this paper was published in[9].

2. AMPLITUDE OF TWO-PHOTON PROCESSES

The amplitude of a two-photon process in the nonrelativistic limit and in the dipole approximation has the following form[1)] (cf., e.g.,[10]):

$$U_{i\to f} = 2\pi\alpha\sqrt{\omega_i\omega_f}\,\langle f|\,(\mathbf{r}\mathbf{e}_f)\,C_{E_i\pm\omega_i}(\mathbf{r},\mathbf{r}')\,(\mathbf{r}'\mathbf{e}_i) + (\mathbf{r}\mathbf{e}_i)\,G_{E_i\pm\omega_f}(\mathbf{r},\mathbf{r}')\,(\mathbf{r}'\mathbf{e}_f)\,i\rangle|, \tag{1}$$

where e and ω are the polarization vector and the photon frequency, E_i is the energy of the atomic level,

$$G_E(\mathbf{r},\mathbf{r}') = \sum_n \frac{\psi_n(\mathbf{r})\psi_n^*(\mathbf{r}')}{E_n - E} \tag{2}$$

is the Green's function of the Schrödinger equation for the hydrogen atom; the sum in (2) extends over all the discrete states and over the continuous spectrum.

The function $G_E(\mathbf{r}, \mathbf{r}')$ can be obtained in explicit form by expanding it in a series in spherical functions

$$G_E(\mathbf{r},\mathbf{r}') = \sum_{lm} g_l(E; r, r')\,Y_{lm}(\mathbf{r}/r)\,Y_{lm}{}^*(\mathbf{r}'/r'), \tag{3}$$

where the radial Green's function g_l satisfies the equation

$$\left\{\frac{1}{2m}\left[\frac{1}{r^2}\frac{d}{dr}\left(r^2\frac{d}{dr}\right) - \frac{l(l+1)}{r^2}\right] + \frac{\alpha Z}{r} + E\right\} g_l(E; r, r') = \frac{\delta(r-r')}{rr'} \tag{4}$$

and is a symmetrical function of r and r′.

A solution of (4), satisfying the necessary boundary conditions ($g_l \sim r^l$ as $r \to 0$; $g_l \to 0$ as $r \to \infty$) can be

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[1)] We use a system of units in which $c = \hbar = 1$ and $e^2 = \alpha = 1/137$.

constructed from the fundamental solution of the homogeneous equation corresponding to (4):

$$g_l(E;r,r')=-\frac{\Gamma(1+l-\nu)}{\Gamma(2l+2)}\frac{ma\nu}{rr'}M_{\nu,\,l+1/2}\left(\frac{2r_<}{a\nu}\right)W_{\nu,\,l+1/2}\left(\frac{2r_>}{a\nu}\right).\qquad(5)$$

Here M and W are the known Whittaker functions, $r_>(r_<)$ is the larger (smaller) of the quantities r and r′, a is the Bohr radius, and $\nu = \alpha Z\sqrt{-m/2E}$ is the "principal quantum number" in the virtual state.

It is seen from the properties of the Γ functions that the radial function (5), together with the complete Green's function (3), has simple poles at the points $\nu = n \geq l + 1$, where n is an integer. Using the relations[11]

$$M_{n,\,l+1/2}(z) = z^{l+1}e^{-z/2}{}_1F_1(-n+l+1,\ \ 2l+2,\ z),$$

$$W_{n,\,l+1/2}(z)=(-1)^{n-l-1}\frac{\Gamma(n+l+1)}{\Gamma(2l+2)}z^{l+1}e^{-z/2}{}_1F_1(-n+l+1,2l+2,z),$$

where ${}_1F_1$ is the confluent hypergeometric function, we can readily show that the residues at these poles are equal to the product of two eigenfunctions of the Schrödinger operator for the hydrogen atom, corresponding to the quantum numbers (n, l, m). The dependence of g_l on r and r′ in terms of the combination

$$r_> = {}^1\!/_2(r+r'+|r-r'|),\qquad r_< = {}^1\!/_2(r+r'-|r-r'|)$$

is not convenient, for its use in the calculation of the transition amplitude makes it necessary to deal with indefinite integrals of Whittaker functions. As will be shown later, it is possible to obtain an exact analytic expression for the bound-bound transition amplitudes by using for the product of the Whittaker functions entering in (5) the following integral representation[12]

$$M_{\nu,\,l+1/2}\left(\frac{2r_<}{a\nu}\right)W_{\nu,\,l+1/2}\left(\frac{2r_>}{a\nu}\right)=\frac{2\sqrt{rr'}}{a\nu}\frac{\Gamma(2l+2)}{\Gamma(1+l-\nu)}\qquad(6)$$

$$\times\int\limits_0^\infty dx\exp\left[-\frac{r+r'}{a\nu}\operatorname{ch}x\right]\left(\operatorname{cth}\frac{x}{2}\right)^{2\nu}I_{2l+1}\left(\frac{2\sqrt{rr'}}{a\nu}\operatorname{sh}x\right),$$

where I is a Bessel function of imaginary argument. The integral (6) exists only when $\operatorname{Re}\nu < l + 1$. This, however, cannot limit the applicability of (6) to the calculation of the amplitudes of such transitions when this condition is not satisfied. In fact, if the calculation yields an analytic expression for the amplitude, this expression can be continued to the region $\operatorname{Re}\nu \geq l + 1$ on the basis of the analyticity of the S matrix with respect to energy.

Recognizing that the radial functions of the hydrogen atom $R_{nl}(r)$ are in the form of a product of a polynomial by an exponential, we obtain after substituting (3), (5), and (6) in (2)

$$Q_{km}(\beta,\beta',l)=\int\int\limits_0^\infty\int dx\,dr\,dr'\,r^{k+5/2}r'^{m+5/2}\exp\left\{-\frac{1}{a\nu}\right.\qquad(7)$$

$$\left.\times\,[\beta r+\beta' r'+(r+r')\operatorname{ch}x]\right\}\left(\operatorname{cth}\frac{x}{2}\right)^{2\nu}I_{2l+1}\left(\frac{2\sqrt{rr'}}{a\nu}\operatorname{sh}x\right),$$

where k and m are integers larger than or equal to the values of the orbital angular momentum of the electron in the initial and final states, respectively, $\beta = \nu/n_i$, and $\beta' = \nu/n_f$.

The minimum possible values of k and m are $l - 1$ (for dipole transitions). The integrals containing r and r′ raised to powers higher than $l - 1$ can be obtained by differentiating $Q_{l-1,\,l-1}$ with respect to β or β′. The exponent 5/2 in Q is the result of the use of formula (1). However, the result can sometimes be obtained in a shorter manner, by using in (1) the known relation between the matrix elements of the momentum and the coordinates $p_{nn'} = im(E_n - E_{n'})r_{nn'}$. In this case, the smallest exponent of r and r′ will be $l - 1 + 3/2$. Thus, all the quantities encountered in $Q_{km}(\beta, \beta', l)$ can be obtained from the following integral, which can be readily calculated:

$$I_l(\beta,\beta')=\int\int\limits_0^\infty\int dx\,dr\,dr'(rr')^{l+1/2}\exp\left\{-\frac{1}{a\nu}[\beta r+\beta' r'+(r+r')\operatorname{ch}x]\right\}$$

$$\times\left(\operatorname{cth}\frac{x}{2}\right)^{2\nu}I_{2l+1}\left(\frac{2\sqrt{rr'}}{a\nu}\operatorname{sh}x\right)=a\nu\int\limits_0^\infty\int dx\,dr\frac{(\operatorname{sh}x)^{2l+1}\left(\operatorname{cth}\frac{x}{2}\right)^{2\nu}}{(\beta+\operatorname{ch}x)^{2l+2}}$$

$$\times\,r^{2l+1}\exp\left\{-\frac{r}{a\nu}\frac{1+\beta\beta'+(\beta+\beta')\operatorname{ch}x}{\beta+\operatorname{ch}x}\right\}$$

$$=\frac{2^{2l+1}(2l+1)!\,(a\nu)^{2l+3}}{(l+1-\nu)[(1+\beta)(1+\beta')]^{2l+2}}{}_2F_1\left(2l+2,l+1-\nu;l+2-\nu;\right.$$

$$\left.\frac{(\beta-1)(\beta'-1)}{(\beta+1)(\beta'+1)}\right)\qquad(8)$$

${}_2F_1$ is a hypergeometric function. This formula yields an analytic expression for the amplitude of any two-photon bound-bound transition.

It is easy to verify that (8) also makes it possible to calculate transitions of arbitrary multipolarity. Indeed, for 2^L-pole transitions the exponent of r in (7) equals $k + L - 1 + 3/2$, and it follows from the selection rules that $(k + L)_{\min} \leq l$. Thus, the minimum exponent for arbitrary L also equals $l + 1/2$.

3. COHERENT AND RAMAN SCATTERING OF PHOTONS BY A HYDROGEN ATOM

Let us consider the application of formula (8) to the scattering of light by a hydrogen atom. Separating the angular parts in (1), we have after summing over the final magnetic quantum numbers of the electron and averaging over the initial ones the following expression for the cross section of the transition $n_i l_i \rightarrow n_f l_f$:

$$d\sigma=\alpha^2\sum_{J=0,\,2}A_JP_J(\cos\theta)\,\omega_i\omega_f^3d\Omega_f,\qquad(9)$$

$$A_J=[(J100|10)]^2\sum_{\lambda\lambda'}[(2\lambda+1)(2\lambda'+1)]^{1/2}\{W(1\lambda1\lambda';l_iJ)$$

$$\times W(1\lambda1\lambda';l_fJ)[M_\lambda(\nu_1)M_{\lambda'}^*(\nu_1)+M_\lambda(\nu_2)M_{\lambda'}^*(\nu_2)]$$

$$+\,2X(1l_i\lambda;\ 1\lambda'l_f;\ J11)\operatorname{Re}[M_\lambda(\nu_1)M_{\lambda'}^*(\nu_2)]\},$$

where $\cos\theta = (\mathbf{e}_i, \mathbf{e}_f)$, P_J is a Legendre polynomial, W and X are the Racah and Fano coefficients, and

$$M_\lambda(\nu)=(l_f100|\lambda0)(\lambda100|l_i0)$$

$$\times\int\int\limits_0^\infty dr\,dr'(rr')^3R_{n_fl_f}(r)R_{n_il_i}(r')g_\lambda\left(-\frac{m(Z\alpha)^2}{2\nu^2};r,r'\right).$$

Here and throughout

$$\nu_1=Z\alpha[-m/2(E_i+\omega_i)]^{1/2},\quad \nu_2=Z\alpha[-m/2(E_i-\omega_f)]^{1/2}.$$

Formulas (8) and (9) make it possible to calculate the scattering cross sections for concrete transitions.

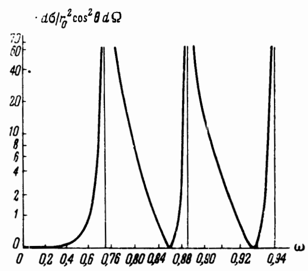

FIG. 1

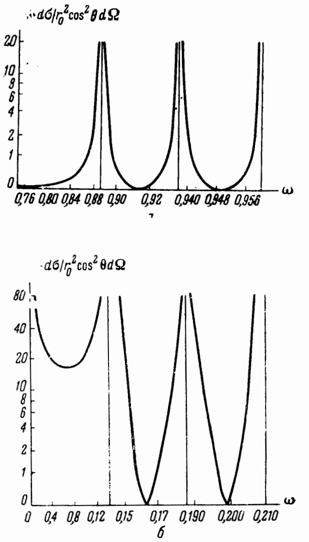

FIG. 2

FIG. 1. Cross section of coherent scattering from 1s level. Ordinates—the quantity $d\sigma/r^2_0\cos^2\theta d\Omega$ in a logarithmic scale. Abscissas—photon frequency ω in units of Ry Z^2. For convenience, a separate scale is used for each interval between poles.

FIG. 2. Raman scattering of light for the 1s → 2s transition (a) and for the 2s → 1s transition. The scales are the same as in Fig. 1. In the case of Raman scattering, just as in the case of coherent scattering, there are frequencies at which the cross section of the process vanishes.

a) Coherent scattering from ns levels (n = 1, 2):

$$d\sigma = r_0^2 \left| \sum_{i=1}^{2} \frac{2(n\nu_i)^2}{(2-\nu_i)(n+\nu_i)^2} \times {}_2F_1\left(1, -1-\nu_i; 3-\nu_i; \left(\frac{n-\nu_i}{n+\nu_i}\right)^2\right) - 1 \right|^2 \cos^2\theta\, d\Omega, \tag{10}$$

where r_0 is the classical radius of the electron.

Figure 1 shows a plot of the scattering on the 1s level, calculated from (10). It is interesting to note that at definite frequencies the scattering cross section drops to zero, i.e., there exist frequencies of "absolute transparency" of atomic hydrogen.

b) Coherent scattering from the 2p shell:

$$\begin{aligned}
d\sigma &= \alpha^2[A_0 + A_2P_2(\cos\theta)]\omega^4 d\Omega,\\
A_0 &= |M_0(\nu_1)|^2 + |M_0(\nu_2)|^2 + |M_2(\nu_1)|^2 + |M_2(\nu_2)|^2\\
&\quad + \operatorname{Re}\{{}^2/_3 M_0(\nu_1)M_0^*(\nu_2) + {}^1/_3 M_2(\nu_1)M_2^*(\nu_2)\\
&\quad + (2\sqrt{5}/3)[M_0(\nu_1)M_2^*(\nu_2) + M_0(\nu_2)M_2^*(\nu_1)]\},\\
A_2 &= {}^7/_{10}[|M_2(\nu_1)|^2 + |M_2(\nu_2)|^2] + 2\operatorname{Re}\{{}^2/_3 M_0(\nu_1)M_0^*(\nu_2)\\
&\quad + {}^{190}/_9 M_2(\nu_1)M_2^*(\nu_2) + (2/\sqrt{5})[M_0(\nu_1)M_2^*(\nu_1) + M_0(\nu_2)M_2^*(\nu_2)]\\
&\quad + (1/3\sqrt{5})[M_0(\nu_1)M_2^*(\nu_2) + M_0(\nu_2)M_2^*(\nu_1)]\},\\
M_0(\nu) &= \frac{8ma^4\nu^2}{3\sqrt{3}(1-\nu^2)(2+\nu)^2(2-\nu)(4-\nu)}\left[\frac{5\nu^4}{2+\nu} \times {}_2F_1\left(1, -2-\nu; 4-\nu; \left(\frac{2-\nu}{2+\nu}\right)^2\right) + 29\nu^4 - 29\nu^3 - 204\nu^2 + 30\nu + 180\right],\\
M_2(\nu) &= \frac{160ma^4\nu^2}{3\sqrt{15}(2-\nu)^2(2+\nu)^2}\left[\frac{8\nu^4}{(2+\nu)^2(3-\nu)} + {}_2F_1\left(1, -2-\nu; 3-\nu; \left(\frac{2-\nu}{2+\nu}\right)^2\right) + 12 - 5\nu^2\right].
\end{aligned} \tag{11}$$

c) Raman scattering for the 1s ⇄ 2s transition:

$$d\sigma = \frac{2^{13}}{3^8} r_0^2 \left| \sum_{i=1}^{2} \frac{\nu_i^2}{4-\nu_i^2}\left[\frac{3}{1+\nu_i}\,{}_2F_1\left(1, -1-\nu_i; 3-\nu_i; \frac{(1-\nu_i)(2-\nu_i)}{(1+\nu_i)(2+\nu_i)}\right) - 1\right]\right|^2 \frac{\omega_f}{\omega_i}\cos^2\theta\, d\Omega.$$

We note that formula (12) can also be used to determine the probability of two-photon decay of the metastable 2s level with the aid of crossing symmetry. A formula for the decay of the 2s level was obtained earlier in[8], where a plot of the photon spectrum was presented and the total decay probability was calculated. Figure 2 shows a plot of the cross section as calculated by formula (12).

d) Raman scattering for the 1s ⇄ 3d transition:

$$\begin{aligned}
d\sigma &= \frac{3}{5} r_0^2 \frac{\omega_f}{\omega_i} \sum_{J=0,\,2} A_J P_J(\cos\theta)\, d\Omega_f,\\
A_0 &= -(1/\sqrt{15})[|\mathfrak{M}(\nu_1)|^2 + |\mathfrak{M}(\nu_2)|^2] + {}^2/_3\operatorname{Re}[\mathfrak{M}(\nu_1)\mathfrak{M}^*(\nu_2)],\\
A_2 &= {}^2/_5\{\sqrt{{}^7/_{12}}[|\mathfrak{M}(\nu_1)|^2 + |\mathfrak{M}(\nu_2)|^2] + {}^1/_3\operatorname{Re}[\mathfrak{M}(\nu_1)\mathfrak{M}^*(\nu_2)]\},\\
\mathfrak{M}(\nu) &= \nu^4[(1+\nu)(3+\nu)^2(2-\nu)(3-\nu)]^{-1}\\
&\quad \times {}_2F_1\left(1, -1-\nu; 3-\nu; \frac{(1-\nu)(3-\nu)}{(1+\nu)(3+\nu)}\right).
\end{aligned} \tag{13}$$

In conclusion we note that a comparison made for those transitions and spectral regions for which earlier calculations are available[1-3,5,6] are in good agreement with our results.

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