Spin-statistic selection rules for multiphoton transitions in atomic systems

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PLAN

- Motivation
- Landau-Yang theorem
- Spin-Statistic Selection Rules
- Proof of SSSR-1 and examples
- Analogy with exclusion principle for electrons
- SSSR-2 and SSSR-3. Direct evaluation and examples
- Possible experimental tests
- Calculations for H-like ions
- Calculations for helium
- Results and outlooks

MOTIVATION

- Ultra-precise test confirms photons are bosons
  Bose-Einstein-statistics-forbidden two-photon excitation in atom

  D. DeMille, D. Budker, N. Derr and E. Deveney, PRL. 83, 3978 (1999)
  D. English, V. V. Yashchuk, and D. Budker, PRL. 104, 253604 (2010)

- Why photons are bosons?

- Application of Landau-Yang Theorem in high-energy physics
  Studying the properties of Higgs-boson, Spin-0 particle

- Spin-Statistic behaviour for multiphoton systems $N_\gamma > 2$

LANDAU-YANG THEOREM (LYT)

L. D. Landau, Dokl. Akad. Nauk SSSR 60, 207 (1948)
C. N. Yang, Phys. Rev. 77, 242 (1950)

Two photons cannot have total angular momentum $J = 1$

This is forbidden by the Bose-Eistein Statistics (BES)

Examples:

1. $Z^0 \nrightarrow 2\gamma$
2. $Ps\,(2^3S_1) \nrightarrow 2\gamma$ annihilation

Charge parity P_C conservation:

$$Z^0: \quad P_C = -1$$
$$Ps: \quad P_C = (-1)^S \;\; S \text{ is spin}$$
$$Ps\,(2^3S_1): \quad P_C = -1$$

Photon system (N_γ - number of photons)

$$P_C = (-1)^{N_\gamma}$$

LYT

In LYT photons are NOT necessarily EQUAL

HOWEVER: For two photons it is always possible to choose the rest frame for the center of inertia (RFCI) (total momentum $\vec{K} = 0$)

In RFCI photons will propagate in opposite directions

$$\vec{k}_1 = -\vec{k}_2$$

and have equal frequencies

$$\omega_1 = \omega_2$$

EXCEPT the case when photons are propagating in the same direction (laser photons!)

DEMATERIALIZATION of the particles (examples 1,2) :
RFCI coincides with the rest frame of this particle (RFP)

$$\text{RFCI}=\text{RFP}$$

ATOMIC PHYSICS

Transitions in He atom or He-like ions

Compare transitions $1s2s\ {}^1S_0 \to (1s)^2\ {}^1S_0 + 2E1$

$1s2s\ {}^3S_1 \to (1s)^2\ {}^1S_0 + 2E1$

a) $W_{2^1S_0 \to 1^1S_0 + 2E1}(\omega_1 = \omega_2) \neq 0$

b) $W_{2^3S_1 \to 1^1S_0 + 2E1}(\omega_1 = \omega_2) = 0$

O. Bely, J. Phys. B 1, 718 (1968)

G. W. F. Drake, G. A. Victor and A. Dalgarno, Phys. Rev. A80, 25 (1969)

Connection with LYT (BES):

D. DeMille, D. Budker, N. Derr and E. Deveney, Phys. Rev. Lett. 83, 3978 (1999)

He atom is not a real neutral system. Therefore only LYT is responsible for the case b)

Confirmation (nonequivalent photons)

$$W_{2^3S_1 \to 1^1S_0 + E1M2, E2M1}(\omega_1 = \omega_2) \neq 0$$

R. W. Dunford, Phys. Rev. A 69, 062502 (2004)

SPIN-STATISTIC SELECTION RULES (SSSR)

SSSR-1

Two equivalent photons involved in any atomic process can have only even values of the total angular momentum J

SSSR-2

Three equivalent dipole photons involved in any atomic process can have only odd values of the total angular momentum $J = 1, 3$

SSSR-3

Four equivalent dipole photons involved in any atomic process can have only even values of the total angular momentum $J = 0, 2, 4$

SSSR-N ?

$N > 3$

$N = N_\gamma - 1$

Connection between SSSR-1 and LYT

LYT: only $J = 1$ is forbidden

SSSR-1: all odd values of J are forbidden
still no contradiction

SSSR-1 is formulated in RFA center of mass (center of inertia) for an atom

When the initial particle (atom) is not dematerializing
$$\mathrm{RFCI} \neq \mathrm{RFA}$$

Definition of the orbital angular momentum $\widehat{\vec{L}}$ depends on the choice of the reference frame. Hence the definition of the total angular momentum $\widehat{\vec{J}} = \widehat{\vec{L}} + \widehat{\vec{S}}$ ($\widehat{\vec{S}}$ is spin) for the photon also depends on the choice of the reference frame.

The choice RFA is more convenient for atomic transitions

FORMAL PROOF OF SSSR-1

Wave functions of the photon in momentum space:

$$\vec{A}^{(s)}_{\omega jm}(\vec{k}) \text{ for absorption} \qquad \vec{A}^{(s)*}_{\omega jm}(\vec{k}) \text{ for emission}$$

ω - frequency, j - total one-photon angular momentum, m - projection, s - type of the photon, electric E or magnetic M, $\vec{k}$ argument in momentum space

Vector component of the photon wave function:

$$\left(\vec{A}^{(s)}_{\omega jm}\right)_i$$

$i = 1, 2, 3$; $i = 1, 2$ - transverse components, $i = 3$ - longitudinal component

E, M photons are transverse (have only $i = 1, 2$)

Parity $P = (-1)^i$ for $s = E$, $P = (-1)^{i+1}$ for $s = M$

$$\widehat{\vec{j}}^{\,2} \left(\vec{A}^{(s)}_{\omega jm}\right)_i = j(j+1) \left(\vec{A}^{(s)}_{\omega jm}\right)_i$$

$$\widehat{j}_z \left(\vec{A}^{(s)}_{\omega jm}\right)_i = m \left(\vec{A}^{(s)}_{\omega jm}\right)_i$$

Two-photon wave function

second-rank tensor symmetrized due to BES for two photons with the same frequency

$$\left(\Phi^{s_1 s_2}_{\omega JM}(\vec{k}_1\vec{k}_2)\right)_{i_1,i_2} = N\sum_{m_1 m_2} C^{JM}_{j_1 m_1 j_2 m_2} \times$$

$$\left[\left(\vec{A}^{(s_1)}_{\omega j_1 m_1}(\vec{k}_1)\right)_{i_1}\left(\vec{A}^{(s_2)}_{\omega j_2 m_2}(\vec{k}_2)\right)_{i_2} + \left(\vec{A}^{(s_1)}_{\omega j_1 m_1}(\vec{k}_2)\right)_{i_2}\left(\vec{A}^{(s_2)}_{\omega j_2 m_2}(\vec{k}_1)\right)_{i_1}\right]$$

J, M - total angular momentum and its projection for two-photon system

N - normalization factor (unessential)

$$\left(\widehat{\vec{j}}_1 + \widehat{\vec{j}}_2\right)^2 \left(\Phi^{(s_1 s_2)}_{\omega JM}\right)_{i_1 i_2} = J(J+1)\left(\Phi^{(s_1 s_2)}_{\omega JM}\right)_{i_1 i_2}$$

$$\left(\widehat{j}_{1z} + \widehat{j}_{2z}\right)\left(\Phi^{(s_1 s_2)}_{\omega JM}\right)_{i_1 i_2} = M\left(\Phi^{(s_1 s_2)}_{\omega JM}\right)_{i_1 i_2}$$

$C^{JM}_{j_1 m_1 j_2 m_2}$ - Clebsch-Gordan coefficient (CGC)

EQUIVALENT PHOTONS

$j_1 = j_2 = j \qquad s_1 = s_2 = s$

changing $m_1 \leftrightarrows m_2$ in the second term in $\left(\Phi^{s\,s}_{\omega JM}(\vec{k}_1\vec{k}_2)\right)_{i_1,i_2}$

$$\left(\Phi^{s\,s}_{\omega JM}(\vec{k}_1\vec{k}_2)\right)_{i_1,i_2} =$$

$$= N\sum_{m_1 m_2}\left(\vec{A}^{(s)}_{\omega j m_1}(\vec{k}_1)\right)_{i_1}\left(\vec{A}^{(s)}_{\omega j m_2}(\vec{k}_2)\right)_{i_2}\left[C^{JM}_{jm_1jm_2} + C^{JM}_{jm_2jm_1}\right]$$

symmetry properties of CGC (for integer j):

$$C^{JM}_{j_2m_2j_1m_1} = (-1)^{j_1+j_2+J}C^{JM}_{j_1m_1j_2m_2}$$

$$\left(\Phi^{s\,s}_{\omega JM}(\vec{k}_1\vec{k}_2)\right)_{i_1,i_2} = N\left[1+(-1)^{2j+J}\right]\sum_{m_1 m_2}\left(\vec{A}^{(s)}_{\omega j m_1}(\vec{k}_1)\right)_{i_1}\left(\vec{A}^{(s)}_{\omega j m_2}(\vec{k}_2)\right)_{i_2}$$

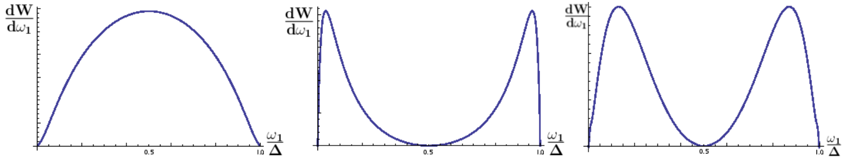
odd values of J are forbidden

SSSR-1. DIRECT EVALUATION. EXAMPLES

QED APPROACH: TWO-PHOTON TRANSITIONS IN HIGHLY CHARGED He-LIKE IONS (HCI)

- Full QED evaluation with neglect of interelectron interaction
- SSSR are the same, but transition probabilities are inaccurate
- The role of interelectron interaction drops down with increase of nuclear charge Z like $1/Z$
- Therefore He-like U: U^{90+} (inaccuracy $\sim 1\%$)

Figure : $\Delta = \omega_1 + \omega_2 = E_i - E_f$



$(1s2s)^1S_0 \to (1s)^{2\,1}S_0 + 2E1$	$(1s2s)^3S_1 \to (1s)^{2\,1}S_0 + 2E1$	$(1s3d)^3D_3 \to (1s)^{2;1}S_0 + 2E2$
0-0 transition	1-0 transition	3-0 transition
$J = 0$ is not forbidden for $\omega_1 = \omega_2$	$J = 1$ is forbidden for $\omega_1 = \omega_2$	$J = 3$ is forbidden for $\omega_1 = \omega_2$

SSSR AS AN EXCLUSION PRINCIPLE FOR PHOTONS: ANALOGY WITH EXCLUSION PRINCIPLE FOR EQUIVALENT ELECTRONS IN ATOMS

Total electron angular momentum allowed by the Fermi-Dirac statistics (FDS) for N_e equivalent electrons within jj coupling scheme

Equivalent electrons: the same principal quantum numbers n_e and the same total one-electron angular momentum j_e

One-electron wave functions in coordinate space

$$\psi_{n_e j_e l_e m_e}(\vec{r}) \text{ - 4-component Dirac spinor}$$

j_e m_e - total one-electron angular momentum and its projection
l_e - orbital one-electron angular momentum
Parity $P = (-1)^{l_e}$

$N_e = 2$. Construction of two-electron wave function:
16-component spinor antisymmetrized according to FDS

$$\psi_{n_{e_1} n_{e_2} J_e M_e}(\vec{r}_1, \vec{r}_2) = N \sum_{m_{e_1} m_{e_2}} C^{J_e M_e}_{j_{e_1} m_{e_1} j_{e_2} m_{e_2}} \times$$

$$\left[\psi_{n_{e_1} j_{e_1} l_{e_1} m_{e_1}}(\vec{r}_1)\psi_{n_{e_2} j_{e_2} l_{e_2} m_{e_2}}(\vec{r}_2) - \psi_{n_{e_1} j_{e_1} l_{e_1} m_{e_1}}(\vec{r}_2)\psi_{n_{e_2} j_{e_2} l_{e_2} m_{e_2}}(\vec{r}_1)\right]$$

J_e M_e - total electron angular momentum and its projection

For equivalent electrons $n_{e_1} = n_{e_2} = n_e$, $j_{e_1} = j_{e_2} = j_e$, $l_{e_1} = l_{e_2} = l_e$
changing $m_{e_1} \leftrightarrows m_{e_2}$ in the second term in square brackets

$$\psi_{n_e n_e J_e M_e}(\vec{r}_1, \vec{r}_2) =$$

$$N\left[1 - (-1)^{2j_e + J_e}\right] \sum_{m_{e_1} m_{e_2}} C^{J_e M_e}_{j_e j_e m_{e_1} m_{e_2}} \psi_{n_e j_e l_e m_{e_1}}(\vec{r}_1)\psi_{n_e j_e l_e m_{e_2}}(\vec{r}_2)$$

Odd values of J_e are forbidden: the same result as for photons. This happens because the sign of the second term in FDS is opposite to BES, but the values of j_e are half-integer

$N_e > 2$. The results can be found in textbooks on atomic theory. They are obtained with the help of Pauli principle

j_e	N_e	J_e
$1/2$	2	0
$3/2$	2	$0, 2$
$5/2$	$2, 4$	$0, 2, 4$
$7/2$	$2, 6$	$0, 2, 4, 6$
	4	$0, 2(2), 4(2), 5, 6, 8$

- We consider only even N_e for comparison with photons
- For N_e even the SSSR similar to photons can be formulated
- Maximum N_e for given j_e: $N_e^{\max} = 2j_e + 1$, $J_e = 0$
- Allowed values of J_e are the same for N_e and $N_e^{\max} - N_e$
- Numbers in parentheses: this J_e is repeated several times
- SSSR similar to photons can be formulated
- Violation of SSSR: $J_e = 5$ for $j_e = 7/2$, $N_e = 4$

  In case of photons SSSR are violated for $j > 1$, $N_\gamma > 2$

FORMAL PROOF OF SSSR-2

For this purpose we use the method of Coefficients of Fractional Parentage (CFP) traditionally employed in atomic theory for construction of the wave functions for systems of equivalent electrons with $N_e > 2$.

Consider the system of 3 equivalent photons:
$\omega_1 = \omega_2 = \omega_3 = \omega$, $j_1 = j_2 = j_3 = j$, $s_1 = s_2 = s_3 = s$

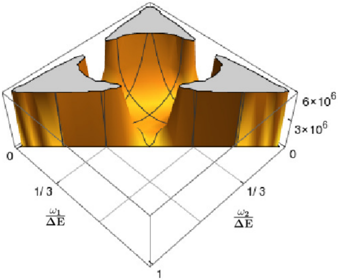
See details in T. Zalialiutdinov, D. Solovyev, L. Labzowsky and G. Plunien, Phys. Rev. A **91**, 033417

This gives a system of equations for CFP which appears to be not resolvable for the values $J = 2$.

SSSR-2. DIRECT EVALUATION. EXAMPLES

The same approach as for SSSR-1: the QED calculation, neglect interelectron interaction.

Object: $U^{90\,+}$; transition $(1s\,2p)^3P_2 \to (1s)^{2\;1}S_0 + 3E1$, 2-0 transition, $J = 2$ is forbidden for $\omega_1 = \omega_2 = \omega_3$



3-dimensional plot of the frequencies distribution in the transition $2^3P_2 \to 1^1S_0 + 3E1$. On the vertical axis transition rate in s^{-1} is plotted; on the two horizontal axes $\omega_1/\Delta E$, $\omega_2/\Delta E$ are plotted, $\Delta E = \omega_1 + \omega_2 + \omega_3 = E_i - E_f$. Coordinates of the lowest point at the bottom of the 'pit': $\omega_1/\Delta E = 1/3$, $\omega_2/\Delta E = 1/3$. Then $\omega_3/\Delta E = 1 - 2/3 = 1/3$.

The value $J = 2$ is forbidden for $\omega_1 = \omega_2 = \omega_3$.

SSSR-3. DIRECT EVALUATION. EXAMPLES

For the direct check of validity of SSSR-1, SSSR-2 the numerical evaluations are not necessary.
It is enough to write down the analytic expression for the transition probability for the photons with the same j, s and set all frequencies equal to each other.
In the same way SSSR-3 can be tested.
Examples:

1) Transition $(1s\,2s)^3S_1 \to (1s)^2\,{}^1S_0 + 4E1$ is forbidden for $\omega_1 = \omega_2 = \omega_3 = \omega_4$, i.e. $J = 1$ is forbidden for the 4-photon system
2) Transition $(1s\,3d)^3D_3 \to (1s)^2\,{}^1S_0 + 4E1$ is forbidden for $\omega_1 = \omega_2 = \omega_3 = \omega_4$, i.e. $J = 3$ is forbidden for the 4-photon system

POSSIBLE EXPERIMENTAL TESTS

- This should be done for neutral He atom, where transitions are in optical region and lasers are available
- 3-photon transitions between fine structure components with account for hyperfine structure H-like ions
- Fixing the initial J_{e_i} and final J_{e_f} momenta for electrons in an atom, we fix the total angular momentum for the photon system
- Choosing the photon number N_γ and defining the laser frequency as $\omega_l = \frac{E_i - E_f}{N_\gamma}$ we choose the laser
- Multipolarity of photons cannot be fixed: since laser light contains all multipolarities, but transitions with $N_\gamma E1$ will be always dominant
- For the certain laser frequencies the atomic vapor should become transparent
- Absorption probabilities with $N_\gamma E1$ photons can be made observable, unlike the spontaneous emission with $N_\gamma E1$ photons

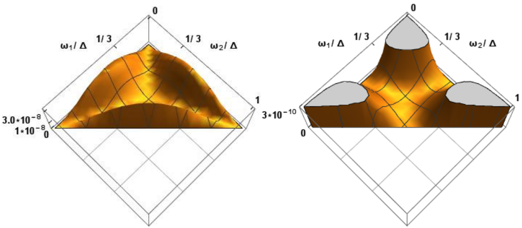
Three-photon transitions in H-like ions

Transitions between fine structure components with the account for hyperfine structure. $I = 3/2$.

Optical range (0.857 eV - 3.27 eV)

a) $2p_{3/2}(F=0) \to 2s_{1/2}(F=2) + 3\gamma(E1)$
b) $2p_{3/2}(F=0) \to 1s_{1/2}(F=2) + 3\gamma(E1)$

a) b)



Ion	Abundance %	$W^{3\gamma}_{vel.}$, s^{-1}	$W^{3\gamma}_{len.}$, s^{-1}	$W^{1\gamma(M2)}$, s^{-1}	ΔE, eV
$^{33}S^{15+}$	0.75	8.944609×10^{-20}	8.944608×10^{-20}	6.406678×10^{-6}	2.992524
$^{35}Cl^{16+}$	93.3	3.404860×10^{-19}	3.404860×10^{-19}	1.917035×10^{-5}	3.817932
$^{39}K^{18+}$	75.8	3.963697×10^{-18}	3.963697×10^{-18}	1.434485×10^{-4}	5.971640

Three-photon transitions in helium

$$2^{3}P_2 \to 2^1S_0 \ \ (1^1S_0) + 3\gamma(E1)$$

Decay channels:

- $2^3P_2 \to n^3S_1 \to n'^3P_1\,[n'^1P_1] \to 1^1S_0 + 3\gamma(E1)$
- $2^3P_2 \to n^3D_1 \to n'^3P_1\,[n'^1P_1] \to 1^1S_0 + 3\gamma(E1)$
- $2^3P_2 \to n^3D_2\,[n^1D_2] \to n'^1P_1 \to 1^1S_0 + 3\gamma(E1)$
- $2^3P_2 \to n^3D_2 \to n'^3P_1\,[n'^1P_1] \to 1^1S_0 + 3\gamma(E1)$

The states admixed by the spin-orbit interaction are placed in the square brackets, n, n' are principal quantum numbers.

$2^{3}P_2 \to 2^1S_0 \ \ (1^1S_0) + 3\gamma(E1)$ transition unlike the $2^{1}P_1 \to 2^1S_0 \ \ (1^1S_0) + 3\gamma(E1)$ transition proceeds only through spin-orbit mixing of the intermediate states.

a) $2^1P_1 \to 2^1S_0 + 3\gamma(E1)$
b) $2^3P_2 \to 2^1S_0 + 3\gamma(E1)$

a) b)

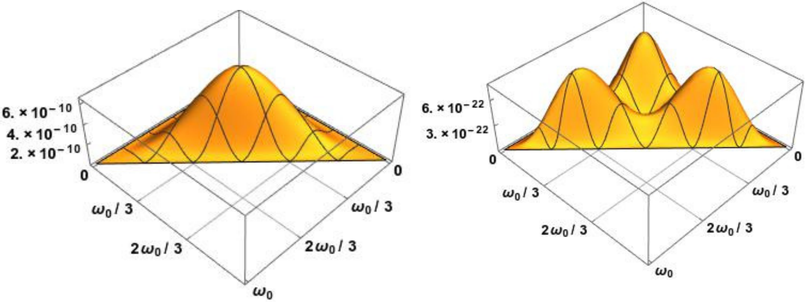


Table : Probabilities for the three-photon $2^3P_2 \to 2^1S_0 + 3\gamma(E1)$, $2^1P_1 \to 2^1S_0 + 3\gamma(E1)$ and one-photon $2^3P_2 \to 2^1S_0 + 1\gamma(M2)$, $2^1P_1 \to 2^1S_0 + 1\gamma(E1)$ transitions in s^{-1}. The number in parentheses indicates the power of ten.

N	$W^{3\gamma(E1)}_{2^3P_2-2^1S_0}$	$W^{3\gamma(E1)}_{2^1P_1-2^1S_0}$	$W^{1\gamma(M2)}_{2^3P_2-2^1S_0}$	$W^{1\gamma(E1)}_{2^1P_1-2^1S_0}$
30	3.727(−27)	4.430(−15)		
50	3.763(−27)	6.651(−15)		
100	3.118(−27)	6.575(−15)		
150	3.117(−27)	6.573(−15)		
500	−	−	1.1953(−8)	1.9746(6)
			1.1952(−8) [1]	

[1] S. A. Alexander, R. L. Coldwell, International Journal of Quantum Chemistry, Vol. 111, 2820-2824 (2011)

SUMMARY

- SSSR were established for multiphoton transitions in atoms
- Direct analytical proof is performed
- Numerical check of SSSR
- QED calculations of three-photon transitions in He-like and H-like ions
- NR calculations of three-photon transitions in helium atom
- Possible experiments are suggested

Thank you for your attention