

SPIN-DEPENDENT SELECTION RULES FOR DIPOLE TRANSITIONS

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Abstract

We have investigated the spin dependent selection rules for dipole transitions in the hydrogen atom by using the solution of Dirac equation . We showed that some transitions allow spin flip which is not possible according to the conventional selection rules. As an example we look at 1s-2p dipole transitions. The percentage of the spin flip rate in (1s-2p) is found to be 56% which is larger than the non-flip rate. The present result is believed to serve a significant help for understanding the recent observation of the spin flip in the excitonic transitions in nanostructures.

1 Introduction

The selection rules for radiative transitions in atoms and molecules are known from the early days of quantum mechanics and can be found in any textbook on quantum mechanics and quantum theory of radiation [1]. In the case of the most frequent electric dipole transitions the selection rules with respect to the total angular momentum $J(J \leftrightarrow J')$ and parity $P(P \leftrightarrow P')$ have the form

$$|\Delta J| \leq 1 \leq J + J' , PP' = -1. \quad (1)$$

In general, the transition probability in atoms strongly depends on the choice of the initial and final atomic states. In the special case of states with spin-orbital (LS) bonds peculiar to the light atoms, the additional selection rules with respect to spin S and orbital angular momentum L and projection of the orbital momentum m are also known:

$$S' - S = 0, \quad |L' - L| \leq 1 \leq L + L', \quad m' - m = 0, \pm 1, \quad (2)$$

where for one-electron atoms like hydrogen, the capital quantum numbers are replaced by small letters ($L \rightarrow l, S \rightarrow s, J \rightarrow j$).

It has been observed recently that the dipole transitions in excitons and some nanostructures manifest the spin-flip effect [2-4]. The main aim of this note is to show that the use of Dirac equation for hydrogen atom in calculation of selection rules allows the spin-flip transitions.

2 The Dirac Hydrogen Atom

Let us begin with the Dirac Hamiltonian of the hydrogen atom [5-7]

$$H_D = \alpha \cdot pc + \beta mc^2 + V(r), \quad (3)$$

where $V(r) = -e^2/r$ is the Coulomb potential, m is the mass of an electron, c is the velocity of light and α and β are the standard Dirac matrices in the Dirac representation

$$\alpha = \begin{pmatrix} 0 & \sigma \\ \sigma & 0 \end{pmatrix}, \quad \beta = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}. \quad (4)$$

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Here the 1's and 0's stand, respectively, for 2×2 unit and zero matrices and the σ is the standard vector composed of the three Pauli matrices $\sigma = (\sigma_x, \sigma_y, \sigma_z)$. Since the Hamiltonian is invariant under rotations, we look for simultaneous eigenfunctions of H_D , $|\mathbf{J}|^2$, and J_z , where

$$\mathbf{J} = \mathbf{L} + \mathbf{S} ; S \equiv \frac{1}{2} \Sigma = \frac{\hbar}{2} \begin{pmatrix} \sigma & 0 \\ 0 & \sigma \end{pmatrix} \quad (5a)$$

and we write

$$J_z = L_z + S_z \quad \text{or} \quad m_j = m_l + m_s \equiv m + m_s \quad (5b)$$

To do this, we remark that the spin operator is diagonal in terms of 2×2 Pauli spin matrices; therefore the angular part should be precisely that of the Pauli two-component theory. Defining $\chi_+ = \begin{bmatrix} 1 \\ 0 \end{bmatrix}$ and $\chi_- = \begin{bmatrix} 0 \\ 1 \end{bmatrix}$ the spin dependent wavefunctions can be written as [8]

$$\begin{aligned} |n, l, m, \uparrow\rangle &\equiv \Psi_{n, j=l+\frac{1}{2}, m_j} = \sqrt{\frac{l+m_j+\frac{1}{2}}{2l+1}} R(r) Y_l^{m_j-1/2} \chi_+ \\ &+ \sqrt{\frac{l-m_j+\frac{1}{2}}{2l+1}} R(r) Y_l^{m_j+1/2} \chi_- \equiv F_1 Y_l^{m_j-1/2} \chi_+ + F_2 Y_l^{m_j+1/2} \chi_- \quad (6) \end{aligned}$$

$$\begin{aligned} |n, l, m, \downarrow\rangle &\equiv \Psi_{n, j=l-\frac{1}{2}, m_j} = -\sqrt{\frac{l-m_j+\frac{1}{2}}{2l+1}} R(r) Y_l^{m_j-1/2} \chi_+ \\ &+ \sqrt{\frac{l+m_j+\frac{1}{2}}{2l+1}} R(r) Y_l^{m_j+1/2} \chi_- \equiv -F_2 Y_l^{m_j-1/2} \chi_+ + F_1 Y_l^{m_j+1/2} \chi_- \quad (7) \end{aligned}$$

3 Developing Selection Rules For The Dirac Hydrogen Atom

If at $t = 0$ the electron is at an initial state state $|i\rangle \equiv |n, l, m, \uparrow\rangle$ given by Eqs. (6) and (7), then at $t > 0$, because of the interaction

with H' , there will be a non-zero transition rate to some other states: $|n', l', m', \uparrow \downarrow\rangle \equiv |f\rangle$ which will be called the final states. The transition probability will be proportional to the square of the matrix element of H' between the initial and the final states:

$$\langle f|H'|i\rangle = \langle n', l', m', \uparrow \downarrow | -\mathbf{d} \cdot \mathbf{E} |n, l, m, \uparrow \downarrow\rangle. \quad (8)$$

To calculate the matrix element in Eq.(8) we do the similar thing as we did earlier. Let us consider two different cases: a) the polarization of the electric field is in x-y plane (along the x- or the y-axis) b) the polarization of the electric field is in z-direction. For the case (a) we need to calculate the matrix elements of the quantities $x \pm iy = r \sin\theta \exp(\pm i\phi)$.

If we separate the matrix elements according to spin orientation for the case (a) we can write:

$$\begin{aligned} & \langle n', l', m', \uparrow \downarrow | -\mathbf{d} \cdot \mathbf{E} |n, l, m, \uparrow \downarrow\rangle \approx \\ & \approx \langle n', l', m', \uparrow | r \sin\theta e^{\pm i\phi} |n, l, m, \uparrow\rangle + \\ & + \langle n', l', m', \downarrow | r \sin\theta e^{\pm i\phi} |n, l, m, \downarrow\rangle + \\ & + \langle n', l', m', \uparrow | r \sin\theta e^{\pm i\phi} |n, l, m, \downarrow\rangle + \\ & + \langle n', l', m', \downarrow | r \sin\theta e^{\pm i\phi} |n, l, m, \uparrow\rangle = \\ & = I_1 + I_2 + I_3 + I_4. \quad (9) \end{aligned}$$

Substitution of Eqs.(6) and (7) in Eq.(9) we get

$$\begin{aligned} I_1 &= \int [F_1' Y_l'^{m_j'-1/2} \chi_+ + F_2' Y_l'^{m_j'+1/2} \chi_-]^* r \sin\theta \times \\ & e^{\pm i\phi} [F_1 Y_l^{m_j-1/2} \chi_+ + F_2 Y_l^{m_j+1/2} \chi_-] d\Omega \approx \\ & \approx \int [Y_l'^{m_j'-1/2}]^* \sin\theta e^{\pm i\phi} Y_l^{m_j-1/2} d\Omega + \\ & + \int [Y_l'^{m_j'+1/2}]^* \sin\theta e^{\pm i\phi} Y_l^{m_j+1/2} d\Omega, \quad (10) \end{aligned}$$

which will be non-zero for

$$\Delta l = \pm 1 \quad \text{and} \quad \Delta m_j = \Delta m_l = \pm 1 \quad (11a)$$

Similarly, the integral I_2 will be non-zero for

$$\Delta l = \pm 1 \quad \text{and} \quad \Delta m_j = \Delta m_l = \pm 1 \quad (11b)$$

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Next we look at the term I_3 :

$$\begin{aligned}
 I_3 &= \int [F'_1 Y_{l'}^{m'_j-1/2} \chi_+ + F'_2 Y_{l'}^{m'_j+1/2} \chi_-]^* r \sin \theta \times \\
 &e^{\pm i\phi} [-F_2 Y_l^{m_j-1/2} \chi_+ + F_1 Y_l^{m_j+1/2} \chi_-] d\Omega \approx \\
 &\approx - \int [Y_{l'}^{m'_j-1/2}]^* \sin \theta e^{\pm i\phi} Y_l^{m_j-1/2} d\Omega + \\
 &+ \int [Y_{l'}^{m'_j+1/2}]^* \sin \theta e^{\pm i\phi} Y_l^{m_j+1/2} d\Omega \quad (12)
 \end{aligned}$$

which will be non-zero for

$$\Delta l = \pm 1 \quad \text{and} \quad \Delta m_j = \Delta m_l + \Delta m_s = \pm 1 + 1 = 0. \quad (13a)$$

Similarly, the integral I_4 will be non-zero for

$$\Delta l = \pm 1 \quad \text{and} \quad \Delta m_j = \Delta m_l + \Delta m_s = \pm 1 - 1 = 0, \quad (13b)$$

where $\Delta m_j = 0$ implies that ($\Delta m_j = \Delta m_l + \Delta m_s = 0$) the sign of Δm_s must have the negative sign of Δm_l . Since for a spin flip transition we have: $\Delta m_s = \mp 1$, then Δm_l should always change sign in the opposite way.

Next we proceed with the case (b) which requires the matrix elements of the quantity: $r \cos \theta$. If we separate the matrix elements according to spin orientation we can write:

$$\begin{aligned}
 &\langle n', l', m', \uparrow \downarrow | -\mathbf{d} \cdot \mathbf{E} | n, l, m, \uparrow \downarrow \rangle \approx \\
 &\approx \langle n', l', m', \uparrow | \cos \theta | n, l, m, \uparrow \rangle + \\
 &+ \langle n', l', m', \downarrow | \cos \theta | n, l, m, \downarrow \rangle + \\
 &+ \langle n', l', m', \uparrow | \cos \theta | n, l, m, \downarrow \rangle + \\
 &+ \langle n', l', m', \downarrow | \cos \theta | n, l, m, \uparrow \rangle = \\
 &= I'_1 + I'_2 + I'_3 + I'_4. \quad (14)
 \end{aligned}$$

Substitution of Eqs.(6) and (7) in Eq.(14) we get:

$$\begin{aligned}
 I'_1 &= \int [F'_1 Y_{l'}^{m'_j-1/2} \chi_+ + F'_2 Y_{l'}^{m'_j+1/2} \chi_-]^* \cos \theta \times \\
 &[F_1 Y_l^{m_j-1/2} \chi_+ + F_2 Y_l^{m_j+1/2} \chi_-] d\Omega \approx
 \end{aligned}$$

$$\approx \int [Y_{l'}^{m'_j-1/2}]^* \cos \theta Y_l^{m_j-1/2} d\Omega + \int [Y_{l'}^{m'_j+1/2}]^* \cos \theta Y_l^{m_j+1/2} d\Omega, \quad (15)$$

which will be non-zero for

$$\Delta l = l' - l = \pm 1 \quad \text{and} \quad \Delta m_j = m'_j - m_j = 0. \quad (16a)$$

Similarly, the integral I'_2 will be non-zero for

$$\Delta l = l' - l = \pm 1 \quad \text{and} \quad \Delta m_j = m'_j - m_j = 0. \quad (16b)$$

Next we consider the matrix elements I'_3 and I'_4 which are involved with the spin flip-flops:

$$\begin{aligned} I'_3 &= \int [F'_1 Y_{l'}^{m'_j-1/2} \chi_+ + F'_2 Y_{l'}^{m'_j+1/2} \chi_-]^* \cos \theta \times \\ &\quad [-F_2 Y_l^{m_j-1/2} \chi_+ + F_1 Y_l^{m_j+1/2} \chi_-] d\Omega \approx \\ &\approx - \int [Y_{l'}^{m'_j-1/2}]^* \cos \theta Y_l^{m_j-1/2} d\Omega + \int [Y_{l'}^{m'_j+1/2}]^* \cos \theta Y_l^{m_j+1/2} d\Omega, \end{aligned} \quad (17)$$

which will be non-zero for

$$\Delta l = \pm 1 \quad \text{and} \quad \Delta m_j = 0. \quad (18a)$$

Similarly, the integral I'_4 will be non-zero for

$$\Delta l = \pm 1 \quad \text{and} \quad \Delta m_j = 0. \quad (18b)$$

To proceed further we substitute Eqs.(9) and (14) in (8):

$$\begin{aligned} \langle f | H' | i \rangle &\approx \langle n', l', m', \uparrow \downarrow | r \sin \theta e^{\pm i\phi} | n, l, m, \uparrow \downarrow \rangle + \\ &\quad + \langle n', l', m', \uparrow \downarrow | r \cos \theta | n, l, m, \uparrow \downarrow \rangle = \\ &= [I_1 + I_2 + I_3 + I_4] + [I'_1 + I'_2 + I'_3 + I'_4] = \\ &= [I_1 + I'_1 + I_2 + I'_2] + [I_3 + I'_3 + I_4 + I'_4], \end{aligned} \quad (19)$$

where the first square bracket corresponds to non-spin flip transitions ($\Delta m_s = 0$) while the second one corresponds to spin flip-flops ($\Delta m_s = \pm 1$). Therefore the selection rules for the first and second term must be considered separately.

For the first square bracket, if we combine Eqs.(11a), (11b), (16a) and (16b); the non-spin flip transitions will occur when

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$$\Delta l = \pm 1 \quad \text{and} \quad \Delta m_j = 0, \pm 1. \quad (20)$$

Similarly combining Eqs.(13a), (13b), (18a) and (18b) in the second square bracket of Eq.(19) the spin flip transitions will be non-zero when

$$\Delta l = \pm 1 \quad \text{and} \quad \Delta m_j = 0. \quad (21)$$

As we stated earlier $\Delta m_j = 0$ implies that ($\Delta m_j = \Delta m_l + \Delta m_s = 0$) the sign of Δm_s must have the negative sign of Δm_l . For a spin flip transition we have: $\Delta m_s = \mp 1$, then $\Delta m_l = -\Delta m_s$ gives $\Delta m_l = \pm 1$ which changes sign in the opposite way.

Now using the addition of angular momentums $\mathbf{J} = \mathbf{J}_1 + \mathbf{J}_2 = \mathbf{L} + \mathbf{S}$ and the Clebsch-Gordon coefficients [9] for an electron the relation between the quantum number (j) and the quantum number (l) can be found as $j = l \mp s$ where (for an electron) the spin quantum number (s) is equal to $(1/2)$ while m_s takes the values ($m_s = \mp s = \mp 1/2$). So Δs is always zero. Then $\Delta j = \Delta l \mp \Delta s = \Delta l$. Combining Eqs. (20) and (21) the selection rules for the Dirac hydrogen atom will be:

$$\Delta j = \mp 1 \quad \text{and} \quad \Delta m_j = 0, \mp 1. \quad (22)$$

As it is well known in the hydrogen atom the potential energy $V(r) = -\frac{e^2}{r}$ is one of the simplest potential in quantum mechanics that can be solved analytically. Although the problem is a two body problem the related wave equation becomes one particle equation after the center of mass motion is separated out. Because of the fact that proton is more massive than electron, we can also assume that the proton is at rest at the origin of the center of mass system. If proton's magnetic moment vector μ_p is in the z-direction, then in the ground state electron will occupy the 1s (spin up) $\equiv |1,0,0, \uparrow\rangle$ state. We will have 6 different spin dependent 2p states which are: $|2,1,0, \uparrow\downarrow\rangle$, $|2,1,1, \uparrow\downarrow\rangle$ and $|2,1, -1, \uparrow\downarrow\rangle$. The matrix elements for these states will be:

$$\begin{aligned} < 2, 1, 0, \uparrow | -\mathbf{d} \cdot \mathbf{E} | 1, 0, 0, \uparrow \rangle = \frac{\sqrt{2}}{3} C, \\ < 2, 1, 0, \downarrow | -\mathbf{d} \cdot \mathbf{E} | 1, 0, 0, \uparrow \rangle = \frac{2}{3} C, \\ < 2, 1, 1, \uparrow | -\mathbf{d} \cdot \mathbf{E} | 1, 0, 0, \uparrow \rangle = 0, \\ < 2, 1, 1, \downarrow | -\mathbf{d} \cdot \mathbf{E} | 1, 0, 0, \uparrow \rangle = -\frac{1}{3} C, \\ < 2, 1, -1, \uparrow | -\mathbf{d} \cdot \mathbf{E} | 1, 0, 0, \uparrow \rangle = -\frac{\sqrt{2}}{3} C, \end{aligned}$$

$$\langle 2, 1, -1, \downarrow | -\mathbf{d} \cdot \mathbf{E} | 1, 0, 0, \uparrow \rangle = 0,$$

where C is a proportionality constant whose detailed calculation is not necessary here.

The related transition probabilities will be $\frac{2}{9}$; $\frac{4}{9}$; 0; $\frac{1}{9}$; $\frac{2}{9}$; 0; respectively, which add to unity: $\frac{2}{9} + \frac{4}{9} + \frac{1}{9} + \frac{2}{9} = 1$.

Next we look at the spin flip rate which is equal to $\frac{5}{9} \cong 56\%$ which is larger than the non-flip rate ($\frac{4}{9} \cong 44\%$).

4 Conclusions

We have investigated the spin dependent selection rules for dipole transitions in the hydrogen atom by using the solution of Dirac equation. We showed that some transitions allow spin flip which is not possible according to the conventional selection rules. As an example we look at 1s-2p dipole transitions. The percentage of the spin flip rate in (1s-2p) is found to be 56% which is larger than the non-flip rate. The present result is believed to serve a significant help for understanding the recent observation of the spin flip in the excitonic transitions in nanostructures.

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5.0.1 References

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Comment on SPIN-DEPENDENT SELECTION RULES FOR DIPOLE TRANSITIONS

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Despite the fact that properties of atomic transitions are well known since the early days of quantum mechanics, the theoretical analysis of peculiarities of the spectroscopic data still attracts a great deal of interest (e.g., see recent monograph [1]).

It is well known that some nanostructures like quantum dots and quantum wells behave like two- or multi-level atoms. For example, an exciton-polariton confined in a quantum well represents an ideal two-level atom, manifesting a number of specific phenomena such as collapse and revival of Rabi oscillations [2, 3, 4]. Similar effects can also be observed in quantum dots [5, 6]. Let us stress that those “artificial atoms” are considered nowadays in the context of design of a hardware for realization of quantum computing.

It has been observed that the dipole transitions in excitons and some nanostructures can manifest spin-flip effects [7, 8].

In the article by M. Saglam *et al*, it is shown that even the conventional Dirac model of hydrogen atom surprisingly allows spin-flip effects. The corresponding rate seems to be quite high (more than 50%).

This result can be a good starting point for further discussion of spin-flip properties of radiative transitions and their possible use as qubits in quantum information technologies.

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