Symmetry and coherent control of the quantum dynamics of Rydberg atoms

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Abstract. Transitions probabilities for highly excited Rydberg states of atoms in electromagnetic traps under the influence of an external electromagnetic fields are calculated. We used the method of groups of dynamical symmetry for hydrogen-like atoms and the technique of relevant coherent states. This allows us to calculate the transition probabilities between atomic levels outside the framework of perturbation theory. The problem of coherent controlling the quantum dynamics of atoms in cavities and some possible applications for quantum information theory are briefly discussed.

1. Introduction

Photonics and quantum optics are now rapidly developing and in many ways the research has moved to the engineering level to create devices that work on the basis of quantum principles. Unique measuring tools have been developed that allow one to operate with single or few atoms and photons. There are fundamentally new objects of research, such as quantum dots in semiconductors, optomechanical devices and artificial atoms created with the help of Josephson rings in physics of superconductors. An active research of traditional objects for quantum optics, such as Rydberg atoms [1-3], excited to very high levels in high-Q resonators is also continuing. At high values of the principal quantum number \( n \), the valence electron has a binding energy, which decreases as \( n^{-2} \) and radius of the Bohr orbit increases as \( n^{2} \) [4]. Rydberg states of atoms and ions are very convenient for observing the quantum effects of interaction of an atom with photons [5]. This is due to the fact that Rydberg states are very well associated with the radiation field, since the amplitudes of transitions between neighboring levels increase as \( n^{4} \). Transitions between neighboring levels with \( n>30 \) are in the millimeter wavelength range, so that resonators with photon modes of small order can be made sufficiently large, providing a big enough time for atom and photon interaction.

Electrons of the inner shells of an atom (ion) are under the influence of a strong electrostatic field of the nucleus \( E_{0} \), and the external fields do not have much significance. A highly excited Rydberg electron moves in a strongly weakened nuclear field \( \sim E_{0} \cdot n^{-4} \), so its quantum state can change significantly in an external electromagnetic field. This was used in recent experiments on the creation of one-atom masers [5].

The unique properties of the Rydberg states make it possible to use the model of hydrogen-like atoms to describe their interaction with an external field and to apply for the calculation of a transition probabilities the symmetry group \( SO(4) \) known for the hydrogen atom. This symmetry group was discovered more than 80 years ago by V. A. Fock and V. Bargmann (see, for example [6,7]). We show here that when calculating transitions between Rydberg states without changing the principal quantum...
number \( n \), this group acts as the group of dynamical symmetry of the Hamiltonian in interaction picture.

The dynamical symmetry group of the Hamiltonian of a quantum system is meant a group whose unitary irreducible representation acts in the Hilbert space of all states of the system [8]. In this case, the Hamiltonian \( \hat{H} \) is assumed to be representable as an operator-valued function of generators \( \hat{A}_1, \ldots, \hat{A}_r \) for the representation of dynamical Lie group \( G \):

\[
\hat{H} = f(\hat{A}_1, \ldots, \hat{A}_r),
\]

where the Hermitian operators \( \hat{A}_1, \ldots, \hat{A}_r \) form a basis of the Lie algebra of the \( M \)-parametric group \( G \):

\[
\left[ \hat{A}_\alpha, \hat{A}_\beta \right] = i C_{\alpha\beta}^\gamma \hat{A}_\gamma,
\]

where \( C_{\alpha\beta}^\gamma \) are the structural constants of a Lie group \( G \), \( (\alpha, \beta, \gamma = 1, \ldots, M) \).

Quantum optics and quantum informatics are a natural field of application of the dynamical group method. One of the main tasks here is to describe the interaction of an external electromagnetic field with matter. If the radiation field is monochromatic, then the transitions between some two levels are significant in the atom (atoms), provided that all other transitions are far from resonance. The dynamical symmetry group of such a system is the unitary unimodular group \( SU(2) \). In the multilevel case, the dynamical group is the group \( SU(N) \), where \( N \) is the number of levels.

For a quantum system with a Hamiltonian linear by the generators of the dynamical group, the evolution operator reduces to the representation operator of the group \( G \):

\[
\hat{U}(t, t_0) = \hat{T}(g(t, t_0)),
\]

where \( g(t, t_0) \) is a trajectory in a group space of \( G \), and its initial element \( g(t_0, t_0) \) is equal to the unit element \( e \) of the group \( G \). The representation operator \( \hat{T}(g(t, t_0)) \) can, in principle, be found exactly using the technique of disentanglement for the evolution operator [9,10]. Knowledge of the dynamical symmetry group \( G \) and the construction of a system of group-theoretical coherent states (CS) [9] allows us to formulate the problem of quantum coherent control of the dynamics of transitions in an external fields. To do this, we should look for a trajectory \( g(t, t_0) \) in \( G \), leading to the generation of some given finite state with the highest possible probability.

We recall that the coherent state [9-11] for representations of the group \( G \) is defined by the formula

\[
\left| CS \right> = \hat{T}(g_Z)\left| \Psi_0 \right>,
\]

where \( g_Z \) is an element of the group \( G \) corresponding to the point of the homogeneous space \( G / G_0 \), that is, the representative of the coset \( g_Z G_0 \) in \( G \), and \( G_0 \) is a subgroup of \( G \), leaving the vector \( \left| \Psi_0 \right> \).

The substitution \( \hat{\Psi}(t) = e^{iZ(t)}\left| Z(t) \right> \) into the temporal Schrodinger equation leads to the search for a trajectory in the CS space. The equations defining trajectories have Hamiltonian form on the Kähler space \( G / G_0 \). The quantum control problem [12] reduces then to the known problem from classical hamiltonian mechanics of finding the optimal trajectory on the homogeneous space of the Lie group \( G \).
2. Rydberg (hydrogen-like) atom in external electromagnetic fields

Let us consider a hydrogen-like atom in a classical electromagnetic field. Its Hamiltonian $\hat{H}$ can be written as the sum of the Hamiltonian of the free atom and the interaction Hamiltonian in the electric dipole approximation:

$$\hat{H} = \hat{H}_A + \hat{H}_I(t),$$

where $\hat{H}_A = \hat{p}^2/(2\mu) - Z_\text{e} e_0^2/|r|$ and $\hat{H}_I = -\hat{d} \cdot \vec{E}(t)$. Here $\hat{p} = -i\hbar\nabla$ is the momentum operator, $\mu$ is the reduced mass, $r$ - the distance between the electron and the nucleus, $Z_\text{e}|e_0|$ is the charge of the nucleus shielded by the inner electron shell, $e_0$ is the elementary charge, $\hat{d}$ is the electric dipole moment operator, and $\vec{E}(t)$ is the electric field strength. It is well known that the symmetry group of a hydrogen-like atom is the group of four-dimensional rotations $SO(4)$. Its generators are the angular momentum operator $\hat{L}$ and the Runge-Lenz operator

$$\hat{A} = -Z_\text{e} e_0^2 r^{-1} + \frac{1}{2\mu}\left(\hat{p} \times \hat{L} - \hat{L} \times \hat{p}\right),$$

which commute with the Hamiltonian $\hat{H}_A$, and whose algebra of commutation relations has the form:

$$\left[\hat{L}_i, \hat{L}_j\right] = i\epsilon_{ijk} \hat{L}_k, \quad \left[\hat{L}_i, \hat{A}_j\right] = i\epsilon_{ijk} \hat{A}_k, \quad \left[\hat{A}_i, \hat{A}_j\right] = -2i\hbar e_0 \epsilon_{ijk} \hat{L}_k, \quad i, j, k = 1, 2, 3.$$

Here $\epsilon_{ijk}$ is the components of antisymmetric Levi-Civita tensor. Next, we consider the bound states of a Rydberg electron and introduce operators $\hat{J}_1, \hat{J}_2$ associated with the angular momentum operator $\hat{L}$ and operator $\hat{N} = (-2\hat{H}_A)^{-1/2} \cdot \hat{A}$ in the following manner [6, 7]: $\hat{J}^{(1,2)} = (\hat{L} \pm \hat{N}) / 2$. Operators $\hat{J}_1, \hat{J}_2$ for different upper indices commute, and for equal ones this operators satisfy the commutation relations of the Lie algebra of the group SU(2), which corresponds to the local isomorphism of $SO(4)$ to direct product of two groups SU(2): $SO(4) \approx SU(2) \times SU(2)$. The fixed value of the principal quantum number $n$ corresponds to a unitary irreducible representation $\hat{J}^{(1,2)}$ of $SO(4)$ group, where $J = J_l \equiv J = (n-1)/2$. For further calculations, it is convenient for us to introduce the eigenvectors of the operators $\hat{J}_z^{(1)}$ and $\hat{J}_z^{(2)}$:

$$\hat{J}_z^{(\sigma)}\{J, m_\sigma\} = m_\sigma \{J, m_\sigma\}, \quad m_\sigma = -J, -J+1, ..., J-1, J; \quad (\sigma = 1, 2).$$

As a result, we obtain a basis for the state vectors of the hydrogen-like atom $\{J, m_1\} \otimes \{J, m_2\} \equiv \{J; m_1, m_2\}$, which are related to the wave functions $\Psi_{n_1, n_2, m}(\xi, \eta, \phi)$ obtained by separating the variables in the Schrödinger Equation in parabolic coordinates, and quantum numbers $m_1, m_2$ are connected [6] with parabolic quantum numbers $n_1, n_2, m : m_1 = (n_1 - n_2 + m)/2, \ m_2 = (n_2 - n_1 + m)/2$. State vectors $\{J; m_1, m_2\}$ are also associated with ordinary eigenvectors $|n; l, m\rangle$, where $l$ is the quantum number of orbital angular momentum (l = 0, 1, ..., n - 1), and $m$ is the quantum number of its projection on the axis z:

$$|J; m_1, m_2\rangle = \sum_{l=0}^{n} |m\rangle \langle l m | J m_1, J m_2\rangle; \quad |m\rangle = \sum_{m_1 + m_2 = m} |J; m_1, m_2\rangle \langle J; m_1, m_2|l m\rangle.$$

Here $\langle j m | j_1, j_2, m\rangle$ are the Clebsch–Gordan coefficients that arise in angular momentum coupling in quantum mechanics [6, 7].
It is well known \[6\] that in the subspace of states with a fixed principal quantum number \(n\) the operator of dipole moment \(\hat{d}\) is proportional to the operator \(\hat{N}\). Using this fact, the interaction Hamiltonian \(\hat{H}_I(t)\) can be represented in the form:

\[
\hat{H}_I(t) = \bar{\omega}^{(1)}(t)\hat{J}^{(1)}(t) + \bar{\omega}^{(2)}(t)\hat{J}^{(2)}(t) = \hat{H}_I^{(1)}(t) + \hat{H}_I^{(2)}(t).
\]

(7)

It is important to note that in this approximation the interaction Hamiltonian \(\hat{H}_I(t)\) commutes with the atomic Hamiltonian \(\hat{A}\). We use here and below the units in which \(\hbar = \mu = |e_0| = 1\), and \(\bar{\omega}^{(1)}(t) = -\bar{\omega}^{(2)}(t) = -\frac{2n}{\sqrt{3a_z}}\hat{E}(t)\), \(\omega^{(1,2)} = (\omega_x^{(1,2)}, \omega_y^{(1,2)}, \omega_z^{(1,2)})\).

Let us rewrite the interaction Hamiltonian using lowering and increasing operators \(\hat{J}^{(1,2)} = \hat{J}_x^{(1,2)} + i\hat{J}_y^{(1,2)}\):

\[
\hat{H}_I(t) = \omega_x^{(1,2)}(t)\hat{J}_x^{(1,2)}(t) + b^{(1,2)}(t)\hat{J}_z^{(1,2)}(t) + \bar{b}^{(1,2)}(t)\hat{J}_z^{(1,2)}(t),
\]

(8)

where \(b^{(1,2)}(t) = \frac{1}{2}(\omega_x^{(1,2)} - i\omega_y^{(1,2)})\).

Since the Hamiltonian is linearly expressed in terms of the generators \(\hat{J}^{(1,2)}\), we seek the evolution operator in the form of the representation operator of the group \(SO(4) = SO(3) \times SO(3) \approx SU(2) \times SU(2)\):

\[
\hat{U}_I(t) = \hat{U}^{(1)}(t) \otimes \hat{U}^{(2)}(t).
\]

(9)

Here

\[
\hat{U}_I^{(1,2)}(t) = \exp(-i\psi_{t,12}^{(1,2)}(t)\hat{J}_z^{(1,2)}(t))\exp(-i\theta_{t,12}^{(1,2)}(t)\hat{J}_y^{(1,2)}(t))\exp(-i\phi_{t,12}^{(1,2)}(t)\hat{J}_z^{(1,2)}(t)) =
\]

\[
= \exp(\alpha_{t,12}(t)\hat{J}_x^{(1,2)}(t))\exp(\beta_{t,12}(t)\hat{J}_y^{(1,2)}(t))\exp(\gamma_{t,12}(t)\hat{J}_z^{(1,2)}(t)).
\]

(10)

In this expression \(\psi, \theta, \phi\) are the Euler angles, and parameters \(\alpha, \beta, \gamma\) are related to them by formulas:

\[
\alpha = -\tan(\theta/2)e^{-i\psi}, \quad \beta = -2\ln\cos(\theta/2) - i(\phi + \psi), \quad \gamma = \tan(\theta/2)e^{-i\phi};
\]

\[
\theta = 2\arctan|\rho|, \quad \phi = -\arg \gamma, \quad \psi = \pi - \arg \alpha.
\]

(11)

Substituting the evolution operator into the nonstationary Schrödinger equation, we obtain a system of first-order nonlinear ordinary differential equations that define the classical dynamics on the \(SO(4)\) group. In the last expressions, the index \(\sigma = 1, 2\) in the variables is omitted to simplify the notation:

\[
\begin{align*}
    i(\dot{\alpha} - \alpha \dot{\beta} - \alpha^2 e^{-\beta} \dot{\gamma}) &= b(t), \\
    i(\dot{\beta} + 2\alpha e^{-\beta} \dot{\gamma}) &= \omega_z(t), \\
    i e^{-\beta} \dot{\gamma} &= \bar{b}(t).
\end{align*}
\]

(11)

Finding the solutions of equations (11) for two values of index \(\sigma\) with initial values \(\alpha(0) = \beta(0) = \gamma(0) = 0\), we determine the transition probabilities by calculation of the matrix elements of the evolution operator that reduce to the product of two Wigner \(D\)-functions for the product of \((2J + 1)\) -dimensional representations of the two groups \(SU(2)\) indicated by the Eq. (9).

For a coherent state of the group \(SU(2) \times SU(2)\):

\[
|Z\rangle = |z_1\rangle \otimes |z_2\rangle,
\]

(12)

we obtain the Riccati equations:

\[
i \dot{z}_\sigma = b^{(\sigma)}(t) + \omega_z^{(\sigma)}(t)z_\sigma - \bar{b}^{(\sigma)}(t)z_\sigma^2, \quad \sigma = 1, 2.
\]

(13)

Here the quantum state
JmJm Jm Jm JJ zz J J z z zJ J JmJm Jm Jm − = + − = ∑

is the well-known CS for SU(2) group [9].

The equations (13) determine the classical dynamics on two homogeneous spaces SU(2)/U(1), each of which is a Bloch sphere under the mapping \( z = -e^{i\theta} \tan(\theta/2) \). As a result, the optimal trajectories in the quantum dynamics control problem can be found on two Bloch spheres.

In the case under consideration, the problem of transitions probability calculation between levels of a Rydberg atom was formally reduced to the dynamics of two qubits in an external field. However, unlike the case of qubits, where the representation of each of the SU(2) groups is two-dimensional, here for every value of \( \sigma \) the representation is defined by a unitary matrix of dimension \( (2J+1) \times (2J+1) \). For the fixed principal number \( n \), the number of sublevels is equal to \( n^2 \), so the use of Rydberg atoms as objects for the purpose of performing mathematical operations and storing data in quantum information schemes seems to be very encouraging.

3. Results of computation

We give examples of the transition probabilities calculation between the Rydberg states of the rubidium ion in the field:

\[
\vec{E} = (E_\perp(t) \cos(\omega t), E_\perp(t) \sin(\omega t), E_\parallel),
\]

which is a superposition of a constant uniform electric field of the trap directed along the z axis, and a field with circular polarization propagating along the z axis.

The transition probability \( W(t) \) calculated below is determined by a general expression of the form:

\[
W(t)\big|_{nm_{\sigma} \to nm_{\sigma}'} = \left\langle nm_{\sigma}, \hat{T}_J(t) \right| nm_{\sigma}' \right\rangle^2 = \left\langle nm_{\sigma}', \hat{T}^{(J+1)}(g(t, t_0)) \right| nm_{\sigma} \right\rangle^2.
\]  

(15)

It is important to note that the transition probability is completely determined by the square of the modulus of the matrix element of the representation operator of the dynamic group of the Hamiltonian and \( g(t, t_0) \in SU(2) \times SU(2) \).

Let us consider two particular cases:

1) the circular field with amplitude \( E_\perp = \text{const} \) and
2) the circular field with \( E_\parallel = E_\perp \exp\left(-\left(t - t_0\right)^2/\tau^2\right) \).

In the case of constant amplitude \( E_\perp \), the evolution operator \( \hat{U}_J(t) = \hat{U}^{(1)}(t) \otimes \hat{U}^{(2)}(t) \), as well as the transition probabilities, is calculated exactly in a coordinate system rotating with the field frequency \( \omega \) around the z axis directed along the field \( \vec{E}_\parallel \). In this coordinate system the interaction Hamiltonian (8) can be reduced to diagonal form independent on time

\[
\hat{H}' = +\Omega_1 \hat{J}^{(1)}_z + \Omega_2 \hat{J}^{(2)}_z.
\]

(16)

Its exact eigenvalues in the basis \( |n; m_1, m_2\rangle \) are equal to \( \Omega(m_1 + m_2) \), and \( \Omega_1 = \Omega_2 \equiv \Omega \), where \( \Omega = \left[\left(\omega_\parallel - \omega_\perp\right)^2 + \omega_\perp^2\right]^{1/2} \). These eigenvalues can be interpreted as the quasienergy levels of a hydrogen-like atom in a periodic external field with a period \( T = 2\pi / \omega \).

As a result, the evolution operator \( \hat{U}_J(t) \) reduces to a simple view:

\[
\hat{U}_J(t) = \left( e^{-i\Omega_1 t_1^2} e^{-i\Omega_2 t_2^2} e^{-i\Omega_1 t_1^2} e^{-i\Omega_2 t_2^2} \right) \otimes \left( e^{-i\Omega_1 t_1^2} e^{-i\Omega_2 t_2^2} e^{-i\Omega_1 t_1^2} e^{-i\Omega_2 t_2^2} \right),
\]

(17)
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\[ \omega_1 = \frac{2n}{3Z} E_1, \quad \omega_\perp = \frac{2n}{3Z} E_\perp, \quad \tan(\theta_{1,2}) = \frac{\omega_1}{\omega_\perp - \omega} \]

\textbf{Figure 1.} Time dependence of the probability \( W(t) \) of rubidium ion in external field (14) for transition \( (n = 21, m_1 = m_2 = 1) \rightarrow (n = 21, m_1 = m_2 = 2) \) \( (\omega = 1, t_0 = 0, \tau = 2, E_\perp = 1, E_\parallel = 0.5) \).

The calculated probabilities of transitions between sublevels with a fixed \( n \) oscillate with the Rabi frequency, a multiple to frequency \( \Omega \).

In the case of Gaussian pulse, numerical solution of equations (11) for different values of the parameters of the model in dimensionless variables were performed. The frequency \( \omega_\parallel \) of the field was put equal to unity, close in value to the value of the Stark splitting of the sublevels. As the control parameter, the field strength of the trap was chosen. The results of one of the calculations are shown in Figures 1 and 2. Comparison of the figures shows that an increase in value of \( E_\parallel \) leads to a significant complication of the time dynamics of the probability of the calculated transition.

\textbf{Figure 2.} Time dependence of the probability \( W(t) \) of rubidium ion in external field (14) for transition \( (n = 21, m_1 = m_2 = 1) \rightarrow (n = 21, m_1 = m_2 = 2) \) \( (\omega = 1, t_0 = 0, \tau = 2, E_\perp = 1, E_\parallel = 1.5) \).

The calculated graphs are in qualitative agreement with some results of the experimental work [1].

4. Conclusions and outlook

The main idea of this paper was to demonstrate how approach of the dynamical group work in problem of Rydberg atom in an external electromagnetic field. It was shown, when the electric dipole transitions between sublevels with a fixed value of the principal quantum number are taken into account, the dynamical group of the Hamiltonian is the symmetry group of the hydrogen atom \( SO(4) \). In a circularly polarized radio-frequency monochromatic field, the evolution operator is exactly calculated in an explicit form. In this case, it is possible also to find exactly the quasi-energy levels of the hydrogen-like atom. In the more general case of a field with a time dependence of the amplitude, the evolution operator is also represented in an explicit form, but to find its parameters it is necessary to use the numerical methods. It is also important to emphasize that the method of dynamical group used here does not require the use of perturbation theory. The probabilities of a transition between
energy levels for a hydrogen-like atom in a classical electromagnetic field of the form (14) are calculated. It is shown that a variation in the cavity field strength $E$ can lead to a significant change in the dependence of the transition probability on time. This allows one to control the transitions between the Rydberg atom levels in cavity.

When modeling the quantum control of the dynamics of a Rydberg atom, starting from the analogy with the problem for the dynamics of a qubit in an external classical field [12], it is necessary to look for the extremum of the functional, which for each of the $SU(2)$ groups reduces to minimizing the expression $\int_0^T \tilde{\omega}^2(t) dt$ where the vectors $\tilde{\omega}^{(1,2)}(t)$ are defined in the expression (7). The problem of light control in optical systems is actively discussed now, see, for example, recent articles [13,14].

In order to perform the calculation of the transition without restriction on the fixed value of the principal quantum number, we need to generalize the $SO(4)$ group used here, to the 15-parameter group $SO(4,2)$, the group of the dynamical symmetry of the hydrogen-like atom taking into account all atomic levels. Then, the problem of calculating probabilities becomes much more complicated, since the interaction Hamiltonian no longer commutes with the free atom Hamiltonian. In addition, we will need to more accurately take into account the influence of the cloud of internal electrons, which in this article has been reduced to shielding the charge of nuclear. The meaning of the so-called quantum defect was taken from experimental works. In addition, to make the calculation using the group approach more realistic, one must take into account the electron spin, and also consider the spin-orbit interaction. When investigating quantum control, one should take into account dissipation effects, which can be described successively using the quantum kinetic equation method. In the future, we hope to publish the results of our research in these areas.

5. References

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