MOLDOVA STATE UNIVERSITY
DOCTORAL SCHOOL OF PHYSICAL, MATHEMATICAL,
INFORMATION AND ENEGINEERING SCIENCES

As a Manuscript
C.Z.U. 530.145+535.14

MÎRZAC, Alexandra

QUANTUM DYNAMICS IN MOLECULAR DIPOLAR SYSTEMS

SUMMARY

Speciality 131.01 - mathematical physics

Author: Alexandra Mîrzac

PhD Supervisor: M.Macovei Macovei Mihai, dr. hab. in phys. and math., prof.

Guidance commission: Pavlina Bouroș, dr. in chemistry, assoc. prof.

Profirie Bardețchi, dr. in phys. and math., assoc. prof.

Ion Cojocaru, dr. in phys. and math., assoc. prof.

CHIȘINĂU, 2022

The thesis was elaborated at the Laboratory of Quantum Photonics at the Institute of Applied Physics.

Author:

Mîrzac Alexandra

PhD Supervisor:

M. Macovei Macovei Mihai, dr. hab. in phys. and math., prof.

PhD Comission:

President of the Comission:	Cliucanov Alexandr, dr. hab. in phys. and math., prof.
Ph.D. supervisor:	Macovei Mihai, dr. hab. in. phys. and math., prof.
Official reviewers:	Bardețchi Profirie, dr. in. phys. and math., assoc. prof.
	Ghiu Iulia, dr. in physics, assoc. prof.
	Tronciu Vasile, dr.hab. in phys. and math., prof.

The thesis defence will take place on the 17 of June 2022, at 14:00, bureau 222, building IV at Moldova State University, Alexe Mateevici str. 60, Chișinău, Moldova.

The doctoral thesis and the summary can be consulted at the Library of the Moldova State University and on the website of the National Agency for Quality Assurance in Education and Research (www.cnaa.md).

Author

Secretary of Doctoral Comission: Ostrovschi Serghei, dr. hab. in phys. and math., prof.

CONTENTS

CONCEPTUAL GUIDELINES OF THE RESEARCH

Actual research status:

The quantum properties of light are the object of studies in quantum optical dynamics. During last decades, experimental and theoretical progress emerged together in explaining light - matter interaction and provided a test-bed of various fundamental aspects of quantum mechanics such us coherence, resonance fluorescence, squeezing, laser cooling and quantum entanglement. Quantum optics has a direct and indirect impact on the development of quantum technologies, whose purpose is to integrate non - classical quantum effects in industrial manufacturing and real feasible quantum computational setups. Quantum properties of light are the foundation of the most promising and potentially challenging quantum technologies.

The study of photon dynamics is the core of quantum optics as the concept of *particles of light* has evolved through various stages of theoretical and technological development. The definition of coherent state of photons as eigenstate of the annihilation operator and the description of photon number statistics and the coherence properties of a laser field was proposed by [1]. One of the main advantages of photons is the implementation of quantum technology operating at temperatures that don’t require cryogenic level and they are hardly affected by the environment. In order to maintain the feasibility and optical setup portability, photonic circuits are manufactured as *photonic chips*, where all the basic elements embedded into a small chip to fulfill the operational stable requirements. The effort in building photonic chips has been improved to integrate photon sources made of nonlinear materials deposited in the chip for on-chip spontaneous parametric down conversion (**SPDC**). Using the photonic chips, various problems of photonics quantum information processing have been proved experimentally including boson sampling [2].

The key concept of quantum optics is the exploration of light matter interaction employing the concept of coherent states and used further to explore high-order coherence of light. Squeezed light - a non-classical sample of light-matter interaction has played a major role in quantum optics development [3]. Quantum optics manages to combine both theoretical and applied technology studies. Thus, squeezed light enables a new type of precision measurement, with application in gravitational wave detection [4] and for noiseless communication [5]. In particular, a squeezed vacuum is generated by spontaneous parametric down conversion (**SPDC**) and is the most frequently used process to generate squeezed states, but also, for example, to generate single and entangled photons. Though isolated two-level systems such as an atom, an ion, a quantum dot or a defect in a diamond are also proper candidates to generate single photons [6].

The quantum optical properties of two-level systems interacting with electromagnetic field constitutes the basis for a wide spectrum of applied problems, including laser science [7], fluorescent spectroscopy [8], nano-imaging [9], design of single photon and multi - photon sources [10, 11] and efficient light emitting devices [12]. It has a certain impact in the development of quantum information theory in the context of coherent qubits control. In particular, single photons are the main tool for quantum key distribution using the quantum informational protocol and for more advanced schemes, required for long-distance quantum communication. The information is usually encoded in the polarization of the photon, and in sources emitting single photons on-demand with high entanglement are of major importance for the implementation of quantum information protocols. In this context, semiconductor quantum dots (**QD**s) are two-level systems perfectly functioning as triggered sources of single photons. Also, quantum dots (**QD**s) are highly advantageous because at resonant excitation they emit single photons with outstanding quantum properties of multiphoton suppression and photon indistinguishability [13].

The theoretical aspects of light-matter interaction often describe the quantum optical system dynamics using the non-diagonal matrix element of the dipole moment operator. However, many systems possess non-zero permanent dipole moment such as polar molecules, an atom polarized by static electric field or an asymmetric quantum dot [14], magnetic dipole atomic transitions detected in rare-earth ions [15, 16] posses a non-zero magnetic dipole different from the case of electric dipole transitions, which are considered zero for atomic eigenstates.

In the majority of studies, two-level systems (**TLS**) are considered to possess a certain spatial parity, or their diagonal dipole matrix has a zero value. However, two-level system with permanent dipole moment or two-level systems (**TLS**) with broken inversion symmetry exhibit appealing properties. Some of the following features have been proved in two-level systems (**TLS**) with broken inversion symmetry: high harmonic generation [17], two-color multiphoton resonances [18], additional resonances in nondegenerate four-wave mixing [19], high reflectivity two-photon phase conjugation [20], new set of peaks detected in the emission spectra [21], revivals and collapse of Rabi oscillations [22, 23], population inversion [24], and enhanced features in the one- and two-photon nonlinear absorption and dispersion [25]. Quantum systems possessing permanent dipole moment are widely explored in the context of multiphoton processes. It was proved that the presence of permanent dipole moment enforces certain changes in multiphoton absorption rates. Dipolar quantum systems can radiate at Rabi frequency and can serve as emitters in the THz frequency region. Also, two-level systems with permanent dipole manifest population inversion in the steady state if they are pumped by two monochromatic laser fields.

Two-level systems exhibit an important non-classical property, namely, the squeezing of the field quadratures in the resonance fluorescence spectrum. In connection with development of highly precision measurement devices and quantum computation, the squeezing of the fluorescent field is an endeavoring research subject of laser driven two and three-level systems. Therefore, the issue of generation non-classical light with enhanced squeezing in dipolar few-level atomic systems is still an interesting area of study. More over the presence of permanent dipole moment (**PDM**) changes straightforward the optical feedback of the system, for example, modification in multiphoton resonant excitation [26, 27]. Regarding the three-level quantum systems possessing a permanent dipole moment (**PDM**), one notes their novel feature to embedded simultaneously the properties the two- and three-level systems as function of the tunable Rabi frequency due to the laser driving due to the presence of permanent dipole moment [2^a,4^a]. Also quantum systems containing a supplementary quantum state reveal a large class of coherent interference effects, as well as the application of three-level qubits in composing and testing quantum protocols and information storage. One of the best experimental examples of three-level systems possessing a permanent dipole moment are semiconductor quantum wells (**QW**), which exhibit quantum interference due to interband transitions and intersubband absorption due to asymmetric structure of the system [28]. The permanent dipole moment within three-level systems influences the levels shifting due to charge redistribution, as well the permanent moments interact with optical fields - an aspect rather less studied.

Consequently, few level atomic systems possessing a non-zero permanent dipole moment can be used for tunable generation of electromagnetic waves. This is especially laborious for frequency ranges where known methods are inefficient, such as terahertz (**THz**) domain [29]. This domain is especially challenging because it lies between radio and optical frequency ranges. Therefore, a search of novel and effective **THz** radiation sources is an emerging task for applied and theoretical quantum optics. Also, multiple quanta processes are considered feasible quantum technologies within few level atomic systems. Thus, the non-resonant multiphoton conversion from optical to microwave and vice versa region is an emerging task.

Objectives of the thesis:

- The demonstration of impact of permanent dipole moment (**PDM**) in the resonance fluorescence, squeezing and total quantum fluctuations spectra of a two-level system.
- The investigation of a laser-pumped three level Λ-type system the upper energy level of which is coupled with a quantum oscillator described by a single quantized leaking mode.
- The demonstration of quantum interference effects induced by emitter's dressed states responsible for flexible lasing and deeper cooling effects.
- The investigation of the possibility of frequency conversion from optical to microwave region, via the resonant pumping of an asymmetrical two-level system incorporated in a quantized single-mode resonator.
- The demonstration of multiphoton features of cavity quantum dynamics containing an asymmetric two-level system using certain multiphoton superposition of generated states.

Research hypothesis:

Two- and three- level systems possessing a non-zero permanent dipole moment interacting with external coherent laser fields exhibit important nonclassical features. Consequently, the permanent non-zero dipole moment becomes an advantageous tool to engineer the properties of novel quantum systems exhibiting novel properties in comparison to the similar systems yet in the absence of permanent dipoles will be demonstrated below.

Analytical methods:

- The rotating wave approximation has been applied in order to neglect the quickly oscillating terms in the Hamiltonian and keep only terms expressing detunings or frequency differences. As well, the Born-Markov approximation has been adopted in order to eliminate the vacuum modes of the electromagnetic field reservoir.
- The method of transformation into the interaction picture was applied in order to remove the time depending terms in the system Hamiltonian using a unitary operator. Rewriting the Schrödinger equation in the interaction picture using the unitary transformation operator, one has modified properly the system Hamiltonian.
- The method of projecting the Bloch equations has been applied in order to describe the evolution of atomic operators, due to the driving and spontaneous emission of the system. An equivalent procedure involved the derivation equations of motion for single or more average operators, which is possible in the Heisenberg picture.
- The method of projecting the master equation into Fock states basis has been applied in order to detect the quantum dynamics of the system, described by a solvable system of coupled equations projected in the system state basis. This methods permits one to derive from the equation of motion the investigated parameters describing the system dynamics.

CONTENT OF THE THESIS

In the first Chapter is presented a review giving a certain insight in the quantum dynamical phenomena detected in few level atomic systems possessing a permanent dipole moment. This chapter highlights that these systems are the proper models suitable for the description of and forecasting new light-matter interactions and propose feasible quantum-optical devices with a wide range of emerging applications. One has over-viewed the two-level system framework altogether with phenomena and applications embedding them. Another specially interesting discussion in this chapter regards the lasing and cooling phenomena in quantum optical systems. The topic of THz waves and multiphoton states generation in quantum optical systems is also considered. Nevertheless, the impact of permanent dipole moment on the quantum dynamical properties of various setups is still neglected and presumed to be zero. Thus, one has contoured the missing block of last decade researches in the field of quantum optics and helped one to define the problem considered in the thesis.

It is obvious to say that modern quantum optics is built on the concept of a few level atom. The most important and general concept introduced in this field is the two-level atom and three-level atom, which is a particular extension of two-level atom model. The physics of two- and three-level systems constitutes the basis for quantum optics and quantum electrodynamics. A considerable impact is observed in the field of photonic quantum technologies, where it enables secure exchange of information via single and multiple photon states. In connection with the development of quantum informatics, squeezing in resonance fluorescence processes of laser-driven few-level molecule possessing permanent dipole has been recognized as crucial resource for quantum information processing. The potential applications with artificial atomic systems have renewed interest towards resonance fluorescence and squeezing of the field quadratures within them.

Furthermore, artificial atomiclike systems exhibit an advantage with respect to engineering of their dipole moments and transition frequencies, which makes them extremely sensitive to ultraweak perturbations and cooling or lasing these systems is of fundamental interest as well. More over, quantum systems with permanent dipoles are shown to generate terahertz waves required in high-precision sensing, imaging, spectroscopy and data communication. From this perspective, the investigation of a laser driven three-level system possessing a non-zero dipole moment and coupled to a quantum oscillator is an emerging research topic, because of possibility to create novel quantum system showing lasing or cooling in a wider range of parameters.

In the second Chapter, one studies squeezing in resonance fluorescence and total quantum fluctuations processes from a two-level system possessing all dipole matrix elements are considered nonzero. We shall begin by defining the system Hamiltonian consisting of all terms describing all types of interactions within the system. In comparison to the similar problem yet in the absence of the permanent dipoles, one will compute the fluorescence spectrum, which will have additional scattered spectral lines and supplementary squeezing regions are found.

We shall consider a two-level system with permanent dipoles interacting with two external coherent laser fields. The first laser is near resonance with the transition frequency of the two-level sample while the second one is close to resonance with the dressed-frequency splitting due to the first laser, respectively, see Fig.1.

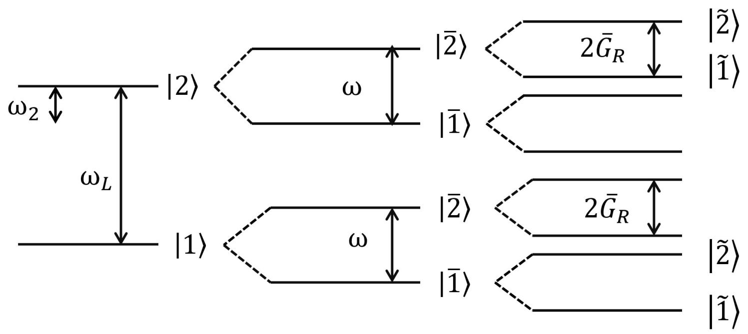


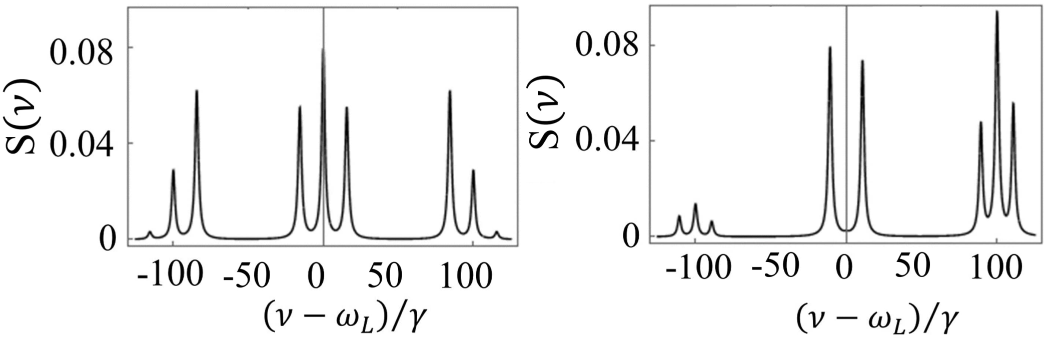
Fig. 1: The energy diagram of a two-level system possessing a permanent dipole. A laser of a moderate intensity with frequency ω_L is interacting with the molecular system, generating the dynamical Stark splitting. The second laser of frequency $\omega = \omega_2$ close to the value of Rabi frequency due to the first laser is leading to transition between double dressed-states. The double dressed-states correspond to the Rabi splitting frequency $2\bar{G}_R$.

Hamiltonian describing the above mentioned setup is developed in the rotating frame at the first laser frequency ω_L and in the dipole approximation is the following:

$$\begin{aligned} H &= \sum_k \hbar\omega_k a_k^\dagger a_k + \hbar\omega_0 S_z + \hbar\Omega_1(S^+ e^{-i\omega_L t} + S^- e^{i\omega_L t}) + \hbar\Omega_2(S^+ + S^-)\cos(\omega t) + \\ &+ \hbar G S_z \cos(\omega t) + \hbar G_1 S_z \cos(\omega_L t) + i\hbar\sum_k (\vec{g}_k \cdot \vec{d})\{a_k^\dagger S^- + a_k S^+\}. \end{aligned} \tag{1}$$

In Hamiltonian (1), the first four components are the free energies of the environmental vacuum modes and molecular subsystems together with the laser-molecule interaction Hamiltonian, respectively. Here, $\Omega_1 \equiv \Omega = dE_1/(2\hbar)$ is the corresponding Rabi frequency with $d \equiv d_{21} = d_{12}$ being the transition dipole moment while E_1 is the amplitude of the first laser field.

The fifth term accounts the interaction of the second laser at frequency ω and amplitude E_2 with the molecular system due to the presence of the permanent dipoles incorporated in G, i.e.,



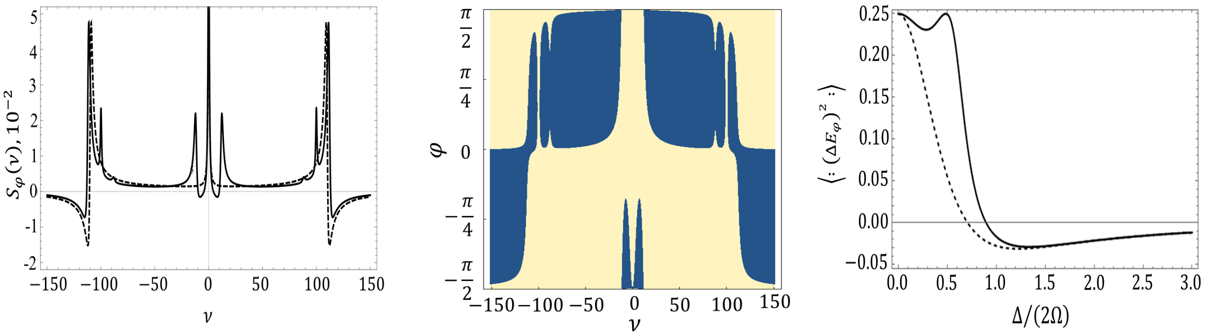
(a)

(b)

Fig. 2: Resonance fluorescence spectra computed for the non-zero permanent dipole moment $G \neq 0$ and the laser detuning ratio over the Rabi frequency:(a) $\frac{\Delta}{2\Omega} = 0$; (b) $\frac{\Delta}{2\Omega} = 0,5$. Here $\frac{\Omega}{\gamma} = 45$ is a parameter representing the ratio between Ω the Rabi frequency and the spontaneous decay rate γ, whereas $\frac{\omega}{\gamma} = 100$ corresponds to the ratio between the frequency of dynamical Stark splitting ω and the spontaneous decay rate.

$G = (d_{22} - d_{11})E_2/\hbar$, while the sixth terms is due to the interaction of the first laser with permanent dipoles. The last term describes the interaction of the molecular subsystems with the environmental vacuum modes of the electromagnetic field reservoir. Further, $\vec{g} = \sqrt{2\pi\hbar\omega k/V}\vec{e}_\lambda$ is the molecule-vacuum coupling strength with $\vec{e}_\lambda$ being the photon polarization vector and $\lambda \in 1, 2$ whereas V is the quantization volume; $\Delta = \omega_{21} - \omega_L$ is the laser field detuning from the molecular transition frequency ω_{21}. The molecule bare-state operators $S^+ = |2\rangle\langle 1|$ and $S^- = [S^+]^\dagger$ obey the commutation relations $[S^+, S^-] = 2S_z$ and $[S_z, S^\pm] = \pm S^\pm$. Here, $S_z = (|2\rangle\langle 2| - |1\rangle\langle 1|)/2$ is the bare-state inversion operator. $|2\rangle$ and $|1\rangle$ are the excited and the ground state of the molecule, respectively $a_k^\dagger$ and a_k are the creation and the annihilation operators of the k_{th} electromagnetic field mode and satisfy standard bosonic commutation relations, namely $[a_k, a_k^\dagger] = \delta_{kk'}$ and $[a_k, a_{k'}] = [a_k^\dagger, a_{k'}^\dagger] = 0$.

We reduce the exponential terms present in the Hamiltonian (1) by transforming it into the Schrödinger picture and applying an affine transformation according to Rotating Wave Approximation (**RWA**). This is required to adopt the presented Hamiltonian (1) of the model to realistic conditions assuming that $\Omega \ll \omega_L \pm \omega$ as well as $\{G, \omega\} \ll \omega_L$ and consequently rapid oscillating terms are dropped off. In this case it is more convenient to describe the system in semi-classical laser-molecule picture in the dressed-states base, due to the first laser pumping: $|2\rangle = -\sin\theta|\bar{1}\rangle + \cos\theta|\bar{2}\rangle$, $|1\rangle = \cos\theta|\bar{1}\rangle + \sin\theta|\bar{2}\rangle$. Additionally after one has defined the new projection base, the new Rabi frequency depends as function of Rabi frequency the first laser pumping and the laser detuning: $\bar{\Omega} = \sqrt{\Omega^2 + \left(\frac{\Delta}{2}\right)^2}$. Therefore the new atomic operators defined in the dressed-state base: $R^+ = |\bar{2}\rangle\langle\bar{1}|$, $R^+ = [R^+]^\dagger$ and the inversion operator $R_z = |\bar{2}\rangle\langle\bar{2}| - |\bar{1}\rangle\langle\bar{1}|$ satisfy the commutation relations: $[R^+, R^-] = 2R_z$ and $[R_z, R^\pm] = \pm 2R^\pm$.



(a) (b) (c)

Fig. 3: Squeezing spectrum $S_\varphi(\nu)$ as function of ν: (a) in the absence of permanent dipole moment $g = 0$ corresponds to the dashed line and in the presence of dipole moment $g = 16$ corresponds to the solid line while the observation angle is $\varphi = -\frac{\pi}{4}$; (b) the projection of squeezing spectrum function in the system of coordinates defined by the observation angle φ and frequency ν. (c) The variance $\langle : \left(\Delta E_\varphi\right)^2 : \rangle$ in units of $|d|^2$ as function of $\frac{\Delta}{2\Omega}$ for $\frac{G}{\gamma} = 0$ (dashed line), $\frac{G}{\gamma} = 16$ (solid line), [1^a]. COLOURED ONLINE

Due to the frequency ω of the second laser another dynamic Stark splitting takes place, we have to simplify once more the system Hamiltonian in the double dressed-state base. We obtain the eigenfunctions of the double dressed - state base as follows: $|\bar{2}\rangle = \sin\bar{\theta}|\tilde{1}\rangle + \cos\bar{\theta}|\tilde{2}\rangle$, $|\bar{1}\rangle = \sin\bar{\theta}|\tilde{1}\rangle + \cos\bar{\theta}|\tilde{2}\rangle$. These eigenfunctions embed the eigenvalues depending on the two - level system parameters as the permanent dipole moment $\bar{G}$ and new generalized Rabi frequency $\bar{G}_R$ projected in the double dressed - state base: $\cos 2\bar{\theta} = \frac{\bar{\Delta}}{\bar{G}_R}$, $\sin 2\bar{\theta} = \frac{\bar{G}}{\bar{G}_R}$, $\cot 2\bar{\theta} = \frac{\bar{\Delta}}{\bar{G}}$, where the generalized Rabi frequency are functions of laser detuning and dipole moment value $\bar{G}_R = \sqrt{\bar{\Delta}^2 + \bar{G}^2}$.The new operators, i.e, $\tilde{R}^+ = |\tilde{2}\rangle\langle\tilde{1}|$,$\tilde{R}^- = [\tilde{R}^+]^\dagger$ and $\tilde{R}_z = |\tilde{2}\rangle\langle\tilde{2}| - |\tilde{1}\rangle\langle\tilde{1}|$, are operating in the double dressed-state picture obeying the following commutation relations:$\left[\tilde{R}^+, \tilde{R}^-\right] = 2\tilde{R}_z$ and $\left[\tilde{R}_z, \tilde{R}^\pm\right] = \pm\tilde{R}^\pm$. Also, the operators projected in the double dressed - state base depend on the laser detuning $\bar{\Delta}$, dipole moment $\bar{G}$ and Rabi frequency $\bar{G}_R$. We substitute the double dressed-state operators, in order to project the system Hamiltonian into the double dressed-state base. Solving eq. (2) in double-dressed state base, one derives the generation and annihilation operators, which contain the expressions for spontaneous decay rates. As well, employing this equation one will derive the average expectation value of the new atomic operators.

$$\begin{aligned}
\frac{d}{dt}\langle Q(t)\rangle &= i\bar{G}_R\langle[\tilde{R}_z, Q]\rangle - \bar{\Gamma}_0\left\{\langle\tilde{R}_z[\tilde{R}_z, Q]\rangle + \langle[Q, \tilde{R}_z]\tilde{R}_z\rangle\right\} \\
&- \bar{\Gamma}_+\left\{\langle\tilde{R}^+[\tilde{R}^-, Q]\rangle + \langle[Q, \tilde{R}^+]\tilde{R}^-\rangle\right\} \\
&- \bar{\Gamma}_-\left\{\langle\tilde{R}^-[\tilde{R}^+, Q]\rangle + \langle[Q, \tilde{R}^-]\tilde{R}^+\rangle\right\}. \qquad (2)
\end{aligned}$$

The master equation (2) contains slowly varying terms in the spontaneous emission damping. Thus, we have assumed that $\bar{G}_R \gg \gamma(\omega_{21})$, with $\gamma(\omega_{21}) = \frac{2d^2\omega_{21}^3}{3\hbar c^3}$, being the single-molecule spontaneous decay rates. With the help of the master equation (2), we obtain the Bloch system of differential equations, describing our sample, which is solved first in stationary case, i.e., we consider that $\frac{d}{dt}\langle\tilde{R}^-(t)\rangle = 0$, $\frac{d}{dt}\langle\tilde{R}^+(t)\rangle = 0$,$\frac{d}{dt}\langle\tilde{R}_z(t)\rangle = 0$.

The phenomenon of resonance fluorescence is the process in which a laser pumped two-level atom scatters photons both coherently and incoherently. Next, we derive the resonance fluorescence for a two-level system possessing a non-zero permanent dipole and explain the phenomena occurring in such theoretical system. The resonance fluorescence spectrum is represented via the terms of the double-correlated functions of the emitted field [30]:

$$S(\nu) = \Phi(r)\int_0^\infty d\tau e^{i(\nu-\omega_L)\tau}\lim_{t\to\infty}\langle S^+(t)S^-(t-\tau)\rangle, \tag{3}$$

where $\Phi(r) = \frac{2d^2\omega_{21}^4}{3r^2c^4}$, r is the distance to the detector.

The spectrum of *resonance fluorescence* describes the light scattered by a two-level system with permanent dipole that is driven by a laser of frequency ω_L and a second laser of frequency ω. The spectrum is sketched in Fig.2(a) and exhibits for a sufficiently large laser intensity three triplets whose width in frequency is of the order of the atomic decay rate $\Gamma_{||}$ and $\bar{\Gamma}_s$, see also [1^a]. The occurrence of the nine lines in the fluorescence spectrum is explained by the two - level system flops at the Rabi frequency $2\bar{G}_R$ between the ground and the excited state. The emission spectrum contains bands at ω_L, $\omega_L \pm \omega$, $\omega_L \pm 2\bar{G}_R$ and side triplets at $\omega_L + \omega \pm 2\bar{G}_R$, $\omega_L - \omega \pm 2\bar{G}_R$. Unlike the spectrum represented in Fig.2(a), the fluorescence spectrum in Fig.2(b) exhibits cancellation of the central line at laser frequency ω_L and the increase in the amplitude of the right sided triplet and the decrease of the left sided triplet. In order to understand the impact of permanent dipole moment on the spectral features of squeezing, we have computed the squeezing spectrum in Fig.3.

In Fig.3(a) is presented the squeezing spectrum for some parameters of interest. Specially, we would like to highlight that squeezing is observed at negative values, which is evident through the solid line in the Fig.3(a) and the dark-blue areas near the zero value of the detector's frequency ν shown in Fig.3(b). In Fig.3(b), we plot the squeezing spectrum for certain parameters of interest. Particularly, squeezing occurs for negative values (dark area in Fig.3(b)) and broader squeezing ranges takes place because of permanent dipoles (see also [1^a]). Especially, squeezing around ν is due to permanent dipoles and will not be be observed in the absence of it. This is a straightforward prove that permanent dipole generates occurrence of new squeezing intervals, which are missing in the squeezing spectra shown by the dashed line.

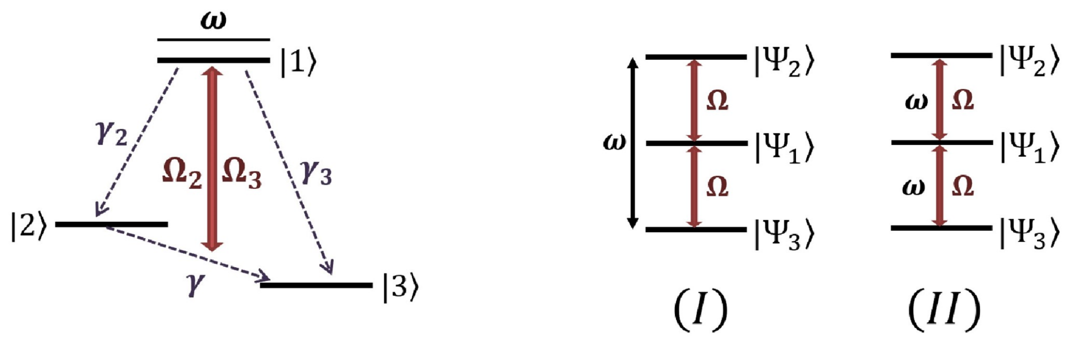
Additionally in Fig.3(c), we depict the normally ordered variance of the radiated $\langle: \left(\mathit{\Delta} E_\varphi\right)^2 :\rangle$, proceeding from the resonance fluorescence processes of laser-pumped two-level systems possessing permanent dipoles. We have found distinct quantum fluctuations features, which are due to permanent dipoles, which is evident from the comparison of the solid and dashed curves, see also [1^a]. The variance curve shown in Fig.3(c) with dashed line was computed for the case when the permanent dipole moment of the two-level system is zero. The presence of the permanent dipole changes the aspect of the normally ordered variance of the radiated field expanding the variance range, though it keeps the same amplitude as compared with dashed line of the normally ordered variance spectra.

In the third Chapter, one investigates the quantum dynamics of a quantum oscillator coupled with most upper state of a three- level Λ-type system. The both transitions of three - level emitter, possessing orthogonal dipole moments, are coherently pumped with a single or two electromagnetic field sources, respectively. One has determined ranges for flexible lasing and cooling phenomena related to the quantum oscillator's degrees of freedom. Due to the asymmetrical decay rates and quantum interference effects, population transfer takes place among relevant dressed states of the emitter's subsystem with which the quantum oscillator is coupled. The most appropriate system can be a nano-mechanical resonator coupled with the most highly energetic state of the three-level emitter place on it. On the other side, if the upper state of the Λ-type system has a permanent dipole it can couple with a cavity electromagnetic field mode oscillating in the terahertz domain, for instance. Furthermore, we demonstrate an effective electromagnetic field source of terahertz photons.

The Hamiltonian describing a quantum oscillator of frequency ω coupled with a laser-pumped Λ-type three-level system, see Fig. 4, in a frame rotating at $\frac{\omega_{12}+\omega_{13}}{2}$, is:

$$H = \hbar\omega b^\dagger b + \frac{\hbar\omega_{23}}{2}(S_{22}+S_{33}) + \hbar g S_{11}(b+b^\dagger) - \hbar \sum_{\alpha\in\{2,3\}} \Omega_\alpha (S_{1\alpha}+S_{\alpha 1}). \tag{4}$$

One has presumed here that as a pumping electromagnetic field source acts a single laser of frequency ω_L irradiating both arms of the emitter or, respectively, two lasers fields $\{\omega_{L1}, \omega_{L2}\}$ each driving separately two transitions of the Λ-type system possessing orthogonal transition dipoles. Supplementary, one has also considered that $\omega_{L1} = \omega_{L2} \equiv \frac{\omega_{12}+\omega_{23}}{2}$, see Fig.4(a). Here $\omega_{\alpha\beta}$ are the frequencies of transitions $|\alpha\rangle \longleftrightarrow |\beta\rangle$ between three-level qubit's, $\{\alpha, \beta \in 1, 2, 3\}$. The terms entering the Hamiltonian (4) have the following meaning, specifically, the first and the second terms describe the free energies of the quantum oscillator and the atomic subsystem. The third term accounts the mutual interaction of the quantum oscillator and the atomic subsystem via the most upper-state energy level with g being the respective coupling strength. The last term corresponds



(a) (b)

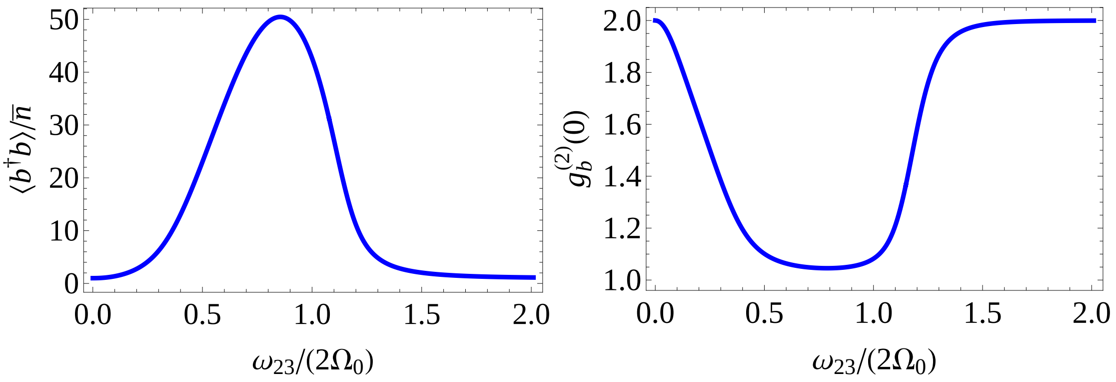
Fig. 4: (a) The schematic set up of the model: a laser-pumped three-level Λ-type system. In this system the upper state, $|1\rangle$ is coupled with a quantum oscillator mode of frequency ω. The oscillator can be associated with a single mode of a nanomechanical resonator containing the three-level emitter. On the other side, if the upper state of the three-level system possesses a permanent dipole then the coupling with an electromagnetic cavity mode occurs in the terahertz ranges of waves, for instance. In this situation, the coupling of the resonator with the lower two levels is insignificant small or, otherwise, the cavity resonant frequency should be out of this resonance with this transition. Additionally, the pumping lasers frequencies are equal to the average transitions frequency of three-level emitter $\frac{\omega_{12}+\omega_{13}}{2}$. Ω_2 and Ω_3 are the frequencies corresponding to laser-qubit coupling strength, ie., the Rabi frequency and γ's are the particular spontaneous decay rates. (b) The semi-classical laser-qubit dressed - state picture where each bare-state level is dynamically split in three dressed states $\Big\{|\Psi_2\rangle, |\Psi_1\rangle, |\Psi_3\rangle\Big\}$. Resonance occur at (I) $\omega = 2\Omega$ or at (II) $\omega = \Omega$, respectively, where Ω is the generalized Rabi frequency, [2^a]. COLOURED ONLINE

to the atom-laser interaction and $\Big\{\Omega_2, \Omega_3\Big\}$ are the corresponding Rabi frequencies associated with a particular driven transitions. Remark that if the upper state of the investigated model contains a permanent dipole moment then the external coherent light sources interact with the upper state as well.

The three-level qubit's operators, $S_{\alpha\beta} = |\alpha\rangle\langle\beta|$, obey the commutation relation $[S_{\alpha\beta}, S_{\beta'\alpha'}] = \delta_{\beta\beta'}S_{\alpha,\alpha'} - \delta_{\alpha',\alpha}S_{\beta'\beta}$, whereas the operators describing the quantum oscillators operators satisfy the following commutation relations $[b, b^\dagger] = 1$ and $[b, b] = [b^\dagger, b^\dagger] = 0$, respectively. In the Born-Markov approximation [31, 32], the quantum dynamics of the proposed complex model can be explored via the following master equation:

$$\begin{aligned}\dot{\rho} + \frac{i}{\hbar}[H, \rho] &= -\sum_{\alpha\in\{2,3\}} \gamma_\alpha [S_{1\alpha}, S_{\alpha}\rho] \\ &- \gamma[S_{23}, S_{32}\rho] - \kappa(1+\bar{n})[b^\dagger, b\rho] - \kappa\bar{n}[b, b^\dagger\rho] + H.c. \qquad (5)\end{aligned}$$

The terms situated on the right side of the eq.(5) corresponds to the emitter's damping due to spontaneous emission as well it accounts the quantum oscillator's damping effects with $\bar{n} =$



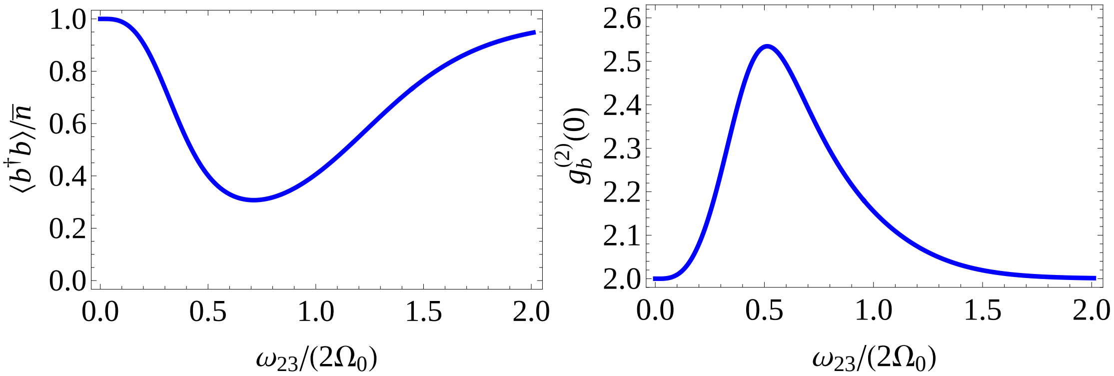
(a)

(b)

Fig. 5: (a) Mean quanta number of the quantum oscillator $\frac{\langle b^{\dagger}b\rangle}{\bar{n}}$ for the situation (I). (b) Presents the second-order correlation function $g_b^{(2)}(0)$ as function of $\frac{\omega_{23}}{2\Omega_0}$ for the situation (I). Here the parameters of interest are $\frac{g}{\gamma_2} = 4, \frac{\gamma_3}{\gamma_2} = 0.1$, $\frac{\gamma}{\gamma_2} = 0, \frac{\kappa}{\gamma_2} = 10^{-3}, \frac{\omega}{\gamma_2} = 20, \frac{\Omega_0}{\gamma_2} = 20$ and $\bar{n} = 1$, [2^a]. COLOURED ONLINE

$\frac{1}{\exp\frac{\hbar\omega}{k_B T}-1}$ being the mean oscillator's quanta number due to the environmental thermostat temperature T. Note that here k_B is the Boltzmann constant, while γ_2 and γ_3 are the spontaneous decay rates corresponding to $|1\rangle \longleftrightarrow |2\rangle$ and $|1\rangle \longleftrightarrow |3\rangle$ transitions, respectively see Fig.4 (a), γ coefficient depicts the spontaneous two - photon decay rate on the $|2\rangle \longleftrightarrow |3\rangle$ transition of the three-level qubit, or the collisional decay rate etc., while κ describes the quantum osccillator's leaking rate, respectively.

The physical phenomena standing behind our model can easier understood if we project the three-level qubit-laser interaction in another dressed-state base: $|1\rangle = \sin\theta|\Psi_1\rangle - \frac{\cos\theta}{\sqrt{2}}\big(|\Psi_2\rangle + |\Psi_3\rangle\big)$,$|2\rangle = \frac{\cos}{\sqrt{2}}|\Psi_2\rangle + \frac{1}{2}(1+\sin\theta)|\Psi_2\rangle - \frac{1}{2}(1-\sin\theta)|\Psi_3\rangle$,$|3\rangle = -\frac{\cos\theta}{\sqrt{2}}|\Psi_1\rangle + \frac{1}{2}(1-\sin\theta)\Psi_2\rangle - \frac{1}{2}(1+\sin\theta)|\Psi_3\rangle$ where $\sin\theta = \frac{\omega_{23}}{2\Omega}$, $\cos\theta = \frac{\sqrt{2}\Omega_0}{\Omega}$ with $\Omega = \sqrt{2\Omega_0^2 + \left(\frac{\omega_{23}}{2}^2\right)}$ being the generalized Rabi frequency whereas $\Omega_2 = \Omega_3 \equiv \Omega_0$. Projecting the Hamiltonian (4) in the new base, one arrives to the corresponding Hamiltonian's expression in the dressed-state picture, i.e, $H = H_0 + H_d + H_1 + H_2$ where $H_0 = \hbar\omega b^{\dagger}b + \hbar\Omega R_z$, $H_d = \hbar g\big(\sin^2\theta R_{11} + \cos^2\theta\frac{(R_{22}+R_{33})}{2}\big)(b+b^{\dagger})$, $H_1 = \hbar\cos^2\theta(R_{32}+R_{23})\frac{(b+b^{\dagger})}{2}$, $H_2 = -\hbar\frac{\sin 2\theta}{2\sqrt{2}}(R_{21}+R_{13}+H.c.)(b+b^{\dagger})$ where $R_z = R_{22} - R_{33}$. Here the dressed-state three - level system operators are $R_{\alpha\beta} = |\Psi_\alpha\rangle\langle\Psi_\beta|$ and they satisfy the same commutation relations as the old ones. In the interaction picture, H_d can be canceled as a fast oscillating term, which can be dropped of the dynamics, while the last two Hamiltonians as: $H_{1I} = \bar{g}(R_{23}e^{2i\Omega t} + H.c.)(b^{\dagger}e^{i\omega t} + H.c.)$ where $\bar{g} = \hbar g\frac{\cos^2\theta}{2}$, $H_{2I} = -\tilde{g}((R_{21}+R_{13})e^{i\Omega t} + H.c.)(b^{\dagger}e^{i\omega t} + H.c.)$ where $\tilde{g} = \hbar g\frac{\sin 2\theta}{2\sqrt{2}}$, are simplified by applying a unitary transformation. According to the above mentioned Hamiltonians one can notice straightforward that the quantum dynamics of the proposed model is determined by two resonant cases, see Fig.4(b), more exactly (I) at $2\Omega = \omega$



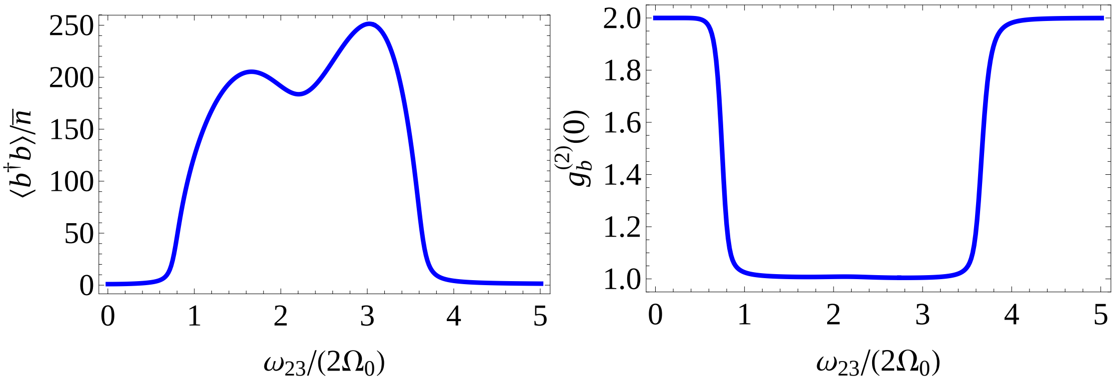
(a) (b)

Fig. 6: (a) Scaled mean quanta number of the quantum oscillator $\frac{\langle b^\dagger b\rangle}{\bar{n}}$ for the situation (I). (b) The corresponding second-order correlation function $g_b^{(2)}(0)$ against the scaled parameter $\frac{\omega_{23}}{2\Omega_0}$ for the situation (I). Here parameters of interest are $\frac{g}{\gamma_3} = 4, \frac{\gamma_2}{\gamma_3} = 0.1$, $\frac{\gamma}{\gamma_3} = 0, \frac{\kappa}{\gamma_3} = 10^{-3}, \frac{\omega}{\gamma_3} = 50, \frac{\Omega_0}{\gamma_3} = 20$ and $\bar{n} = 15$, [2^a]. COLOURED ONLINE

and (II) at $\Omega = \omega$. Consequently, in what follows, we will consider these two cases separately. Thereby, the Hamiltonanian for the first case (I) is: $H = \bar{\delta} b^\dagger b + \bar{g}(R_{32} b^\dagger + b R_{23})$, for the second case (II) is: $H = \tilde{\delta} b^\dagger b - \tilde{g}((R_{12} + R_{31}) b^\dagger + b(R_{21} + R_{13}))$, where $\bar{\delta} = \omega - 2\Omega$ and $\tilde{\delta} = \omega - \Omega$. Note that rapidly oscillating terms in the above Hamiltonians will be dropped off, meaning that $\Omega \gg \{g, \gamma, \gamma_2, \gamma_3\}$, which corresponds to the definition of the secular approximation, according to which the Rabi frequency is much greater then the spontaneous decay rates and the coupling constant of the most energetic level coupled with an electromagnetic cavity.

In the following, we will compare the two cases, i.e., (I) and (II), for the same parameters range and the physical phenomena behind them will be discussed, as well the mechanism behind them. One is going to demonstrate the different mechanisms generating lasing and cooling in the three-level Λ-type system and the novel properties embedded in the model. One is going to present the results for both cases separately in order to avoid confusion and give a distinct interpretation to each particular process.

In the next step, one is presenting the quantum statistics and the mean quanta number is shown in Fig.5. Here, one can observe the maximum value for $\langle b^\dagger b\rangle$ occurs around $\bar{\delta} = 0$, i.e., at the resonance when the quanta's frequency ω is equal to the dressed - state splitting frequency 2Ω due to pumping laser. It is important to mention here that the quanta's statistics is near Poissonian, which means that we have determined lasing regimes in our system. This result is evident from Fig.5(a) and (b). Also, lasing takes place if is satisfied the following condition $\frac{\gamma_3}{\gamma_2} \ll 1$. In this case $\langle R_{22}\rangle > \langle R_{33}\rangle$, this means we have population inversion of the dressed - states, which means we have the lasing effect in our system and evident in Fig.5(a). To avoid any misunderstandings



(a)

(b)

Fig. 7: (a) The mean quanta number of the quantum oscillator $\frac{\langle b^\dagger b\rangle}{\bar{n}}$ for the situation (II). (b) Second-order correlation function $g_b^{(2)}(0)$ versus $\frac{\omega_{23}}{2\Omega_0}$ for the situation (II). Here the parameters of interest are $\frac{g}{\gamma_2} = 4, \frac{\gamma_3}{\gamma_2} = 0.1$, $\frac{\gamma}{\gamma_2} = 0, \frac{\kappa}{\gamma_2} = 10^{-3}, \frac{\omega}{\gamma_2} = 20, \frac{\Omega_0}{\gamma_2} = 20$ and $\bar{n} = 1$, [2^a]. COLOURED ONLINE

via *lasing* we mean *generation of the quantum oscillator's quanta possessing Poissonian statistics*, i.e., $g_b^{(b)}(0) = 1$.

Subsequently, Fig.6(a) and 6(b) brings out the cooling regimes in this system in the context of resonant case (I). This takes place when $\frac{\gamma_2}{\gamma_3} \ll 1$, which means more exactly that $\langle R_{22}\rangle < \langle R_{33}\rangle$ leading to quanta's absorption processes. The minimum value in the mean quanta number followed by an increased second-order correlation function $g_b^{(2)}(0)$ is observed at $\bar{\delta} = 0$, which is the resonance condition noticeable in Fig.6(a) and 6(b).

Here, one can draw some important conclusions regarding the cooling and lasing phenomena mechanisms standing behind resonant case (I). If $\gamma_2 \neq \gamma_3$ and $\gamma = 0$, the first situation (I) corresponds to a two - level system $\{|\Psi_2\rangle, |\Psi_3\rangle\}$ of frequency 2Ω interacting, correspondingly, with a quantum oscillator of frequency ω, we mean here that $2\Omega \approx \omega$. The spontaneous decay functions in both directions, i. e., $|\Psi_2\rangle \longleftrightarrow |\Psi_3\rangle$, with a reciprocal impact on cooling or lasing effects.

In the following, Fig.7(a) shows the mean quanta number of the quantum oscillator in this case (II). The mean oscillator's quanta number, reproduced in Fig. 7(a), exhibits an asymmetric shape, in a certain point of view it is related to a *Fano*-like profile, forecasting the interference effects to exhibit a major role here. Reciprocally, Fig. 7(b) displays the comparable behavior of the second order quanta's correlation function depending on $\frac{\omega_{23}}{2\Omega_0}$ when $\frac{\gamma_3}{\gamma_2} \ll 1$. It is important to mention that one can notice a wide plateau where quanta's statistics is Poissonian at the same time its quantum oscillator's mean quanta numbers range from small to larger values. Thus, we have an evident lasing effect in this theoretical setup.

Correspondingly, Fig.8(a) highlight the cooling regime in the studied system, and for the situa-

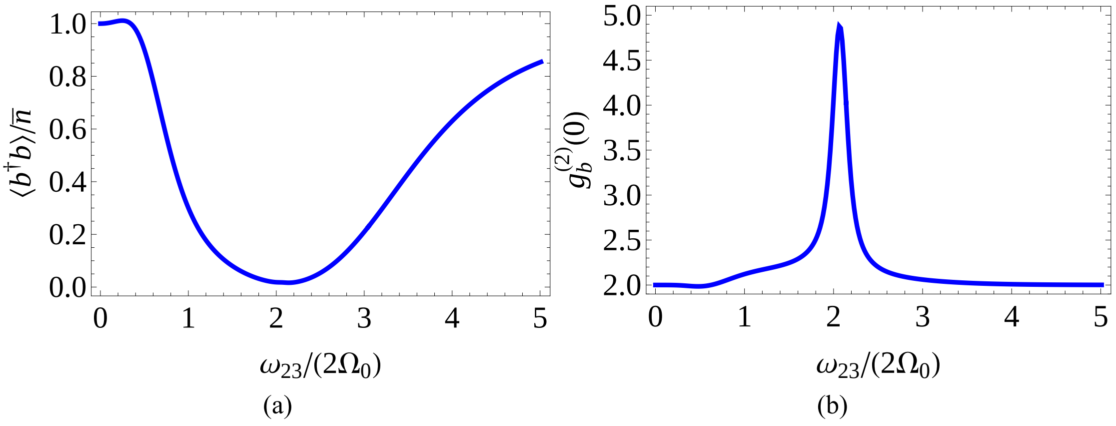


Fig. 8: (a) Scaled mean quanta number of the quantum oscillator for the situation (II) $\frac{\langle b^{\dagger}b\rangle}{\bar{n}}$. (b) The second-order correlation function for the situation (II) $g_b^{(2)}(0)$ versus $\frac{\omega_{23}}{2\Omega_0}$ and the parameters of interest are $\frac{g}{\gamma_3}=4, \frac{\gamma_2}{\gamma_3}=0.1$, $\frac{\gamma}{\gamma_3}=0, \frac{\kappa}{\gamma_3}=10^{-3}, \frac{\omega}{\gamma_3}=50, \frac{\Omega_0}{\gamma_3}=20$ and $\bar{n}=15$, [2^a]. COLOURED ONLINE

tion (II), which takes place when $\frac{\gamma_2}{\gamma_3} \ll 1$. The second-order correlation function increases appropriately, see Fig.8(b), proving enhanced phonon-phonon or photon-photon correlations as function of the model proposed in this chapter. Compared with Fig.6(b) describing the same phenomenon but for the first situation (I), the cooling process is rather improved in the second case (II) at the same time keeping the identical parameters, see Fig. 6(b) and 8(b).

Here, one can observe that according to the resonant case (II) the three-level Λ-type system is similar to an equidistant three-level system $|\Psi_2\rangle \longleftrightarrow |\Psi_1\rangle \longleftrightarrow |\Psi_3\rangle$, where each transition occurs at frequency Ω, interacting as well with the quantum oscillator possessing the frequency ω, however, with $\Omega \approx \omega$. In this circumstance, transitions may emerge via a single oscillator's quanta processes among the dressed-state $|\Psi_2\rangle \longleftrightarrow |\Psi_1\rangle \longleftrightarrow |\Psi_3\rangle$, or, correspondingly involving two-quanta effects among the dressed-states $|\Psi_2\rangle \longleftrightarrow |\Psi_3\rangle$.

In the fourth Chapter, our purpose is the investigation of the multiphoton quantum dynamics of a leaking single-mode quantized cavity field coupled with a resonantly laser pumped two-level system or qubit possessing permanent dipole moment. Most of the well-known frequency conversion investigations involve resonant processes. For this reason, we are going to prove a photon conversion setup exhibiting non-resonant multiphoton effects, respectively. In this context, we are conducting an investigation about frequency down-conversion processes through a resonantly laser-driven emitter possessing a non-zero permanent diagonal dipole moment, $d_{\alpha\alpha} \neq 0$ with $\alpha \in \{1,2\}$ and placed in a quantized resonator presented in Fig.9. The frequencies of the two-level qubit and the single-mode cavity are remarkably different from each other, namely optical and microwave ranges. Consequently, the two-level emitter couples by default to the resonator only through its permanent parallel components of the dipole moment. The cavity's frequency has a different value from the generalized Rabi frequency that arises due to resonant and coherent external pumping of the two-level emitter. The Master Equation analyzing the interaction of

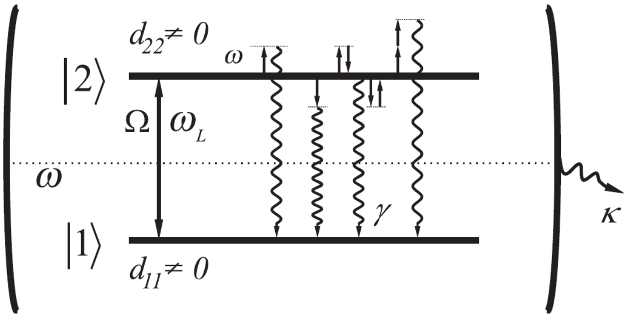


Fig. 9: The scheme of the investigated model. It consists of a coherently pumped two-level system interacting with a single-mode resonator of frequency ω through its non-zero parallel components of the permanent dipole moment, $d_{\alpha\alpha}$, with $\alpha \in \{1,2\}$. In this schematic setup, Ω is the corresponding Rabi frequency due to the off-diagonal dipole moment d_{21} whereas ω_L, the laser frequency is greater then the frequency of the resonantly applied external field, namely $\omega_L \gg \omega$. Whereas g expresses the two- level qubit - resonator coupling strength [6^a].

a two-level qubit, possessing permanent diagonal dipole moment, with a classical coherent electromagnetic field of frequency ω_L as well as with a quantized single mode resonator frequency $\omega \ll \omega_L$ see Fig.9 and is being damped via the corresponding environmental bath in the Born - Markov approximations is:

$$\frac{d}{dt}\rho(t) + \frac{i}{\hbar}[H,\rho] \;=\; -\frac{\gamma}{2}[S^+, S^-\rho] - \frac{\kappa}{2}(1+\bar{n})[b^\dagger, b\rho] - \frac{\kappa}{2}\bar{n}[b, b^\dagger\rho] + H.c. \tag{6}$$

In this master equation (6), γ is the single-qubit spontaneous decay rate, whereas κ is the corresponding boson's leaking mode with $\bar{n} = \left[\exp\left(\frac{\hbar\omega}{k_B T}\right) - 1\right]^{-1}$ is the mean resonator's photon number due to thermal bath temperature T, and k_B is the Boltzmann constant. The two - level qubit might have transition frequency in the optical range of frequencies, while the single - mode cavity frequency is situated in the microwave range of frequencies, respectively. The wavevector of the coherent applied field is perpendicular to the cavity axis. As well, in eq.(6) the bare - state qubit's operators are defined as follows: $S^+ = |2\rangle\langle 1|$ and $S^- = [S^+]^\dagger$ are verifying the commutation relations defined in $SU(2)$ algebra, as follows: $[S^+, S^-] = 2S_z$ and $[S_z, S^\pm] = \pm S^\pm$, where $S_z = \frac{\left(|2\rangle\langle 2| - |1\rangle\langle 1|\right)}{2}$ is the bare state inversion operator. Note that, $|2\rangle$ and $|1\rangle$ are corresponding to the excited and ground state of the qubit, respectively, while $b^\dagger$ and b are the creation and annihilation operator of the electromagnetic field (**EMF**) in the resonator, are satisfying the standard bosonic commutation relation, i.e., $[b, b^\dagger] = 1$, and $[b, b] = [b^\dagger, b^\dagger] = 0$.

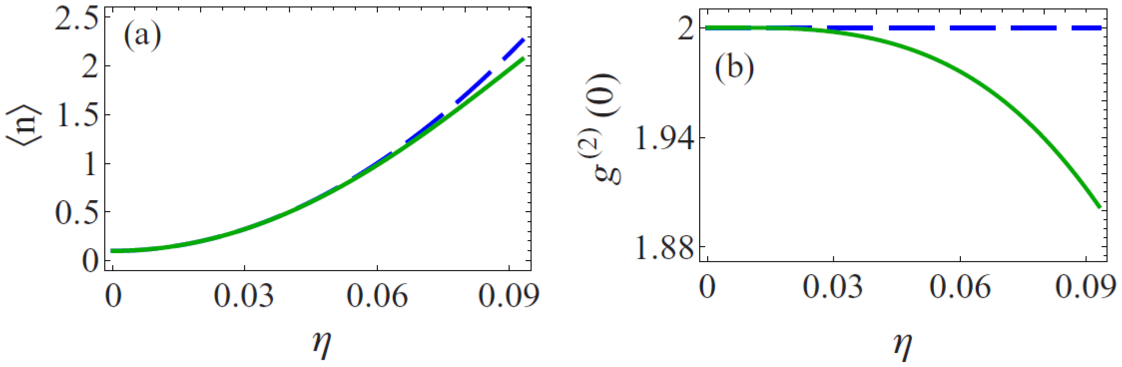


Fig. 10: In the left side (a) of the figure is presented the steady-state mean cavity photon number $\langle n\rangle \equiv \langle \bar{b}^{\dagger}\bar{b}\rangle$ as well as in the right side (b) is presented the second-order correlation function of $\eta = \frac{g}{2\Omega}$. The blue lines are plotted for single-photon processes $N = 1$, while the green ones for two-photon processes, $N = 2$, respectively. Here, $\bar{n} = 10^{-1}$, $\frac{\kappa}{\gamma} = 10^{-3}$ and $\xi = 0$, [6^{a}]. COLOURED ONLINE

The Hamiltonian describing completely the interaction of a two-level system possessing permanent dipoles with an external resonant coherent field as well as with a single-mode resonator, in the dipole and rotating wave approximations, is:

$$\begin{aligned} H &= \hbar\omega b^{\dagger}b + \hbar\omega_{21}S_z - \hbar\Omega\big(S^{+}e^{i\omega_L t} + S^{-}e^{i\omega_L t}\big) + \hbar g_0\big(d_{22}S_{22} + d_{11}S_{11}\big)\big(b^{\dagger} + b\big) \\ &+ \hbar\bar{g}_0\big(S^{+} + S^{-}\big)\big(b^{\dagger} + b\big) - E_L\big(d_{22}S_{22} + d_{11}S_{11}\big)\cos\big(\omega_L t\big). \end{aligned} \tag{7}$$

In this Hamiltonian (7), the first two terms correspond to the free energies of the resonator and the two-level subsystem. The third and the sixth terms of this Hamiltonian (7) describes the interaction of the external laser field with the two-level emitter through its off-diagonal dipole moments d_{21}, $d_{21} = d_{12}$, as well as the diagonal dipole moments d_{22} and d_{11}, correspondingly. The fourth and the fifth terms of the Hamiltonian account for the interactions of the cavity mode with the two-level system va diagonal and off-diagonal dipole moments. Also, E_L is the amplitude of the external driving field, while $g_0 = \sqrt{\frac{2\pi\omega}{\hbar V}}$ where V is the quantization volume, and $\bar{g}_0 = g_0 d_{21}$. $S_{\alpha\alpha}$,$\big\{\alpha = 1, 2\big\}$ are the population operators respectively. One can notice that the fifth's Hamiltonian's term is a rapidly oscillating since ω_L is bigger than the corresponding coupling strength, i. e., $\omega_L \gg \bar{g}_0$ and $\omega_L \gg \omega$, and with respect to this term we have performed the unitary transformation $\bar{U} = \exp\big(i\omega_L S_z t\big)$.

The last term in the Hamiltonian (7) is neglected for the similar reason since $\omega_L \gg \left\{\frac{E_L d_{22}}{\hbar}, \frac{E_L d_{11}}{\hbar}\right\}$ for moderate assumed external pumping strengths. In what follows, we present the diagonalization of Hamiltonian (7) and we have computed the new basis necessary for the further diagonalization: $|2\rangle = -\sin\chi|\bar{1}\rangle + \cos\chi|\bar{2}\rangle$, $|1\rangle = \cos\chi|\bar{1}\rangle + \sin\chi|\bar{2}\rangle$ where $\cos\chi = \frac{\bar{\Delta}}{2\bar{\Omega}}$, $\sin\chi = \frac{\Omega}{\bar{\Omega}}$. We project the

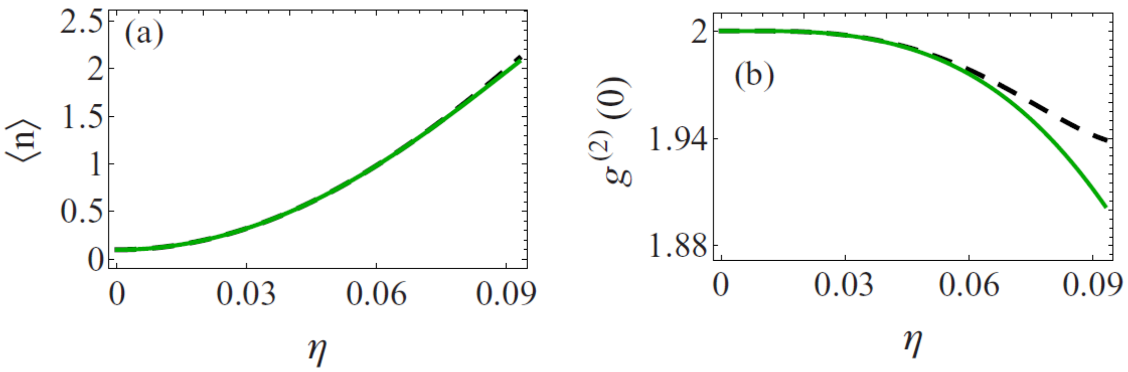


Fig. 11: In the left side (a) of the figure is presented the steady-state mean cavity photon number $\langle n\rangle \equiv \langle \bar{b}^\dagger \bar{b}\rangle$ as well as the right side (b) its second-order correlation function $g^{(2)}(0)$ as a function of $\eta = \frac{g}{2\Omega}$. The green curves are plotted for two-photon processes, $N = 2$, while the black ones are computed for three-photon processes, $N = 3$, respectively and keeping the same parameters as in Fig.10, [6^a]. COLOURED ONLINE

Hamiltonian (7) in the new base using old basis operators presented via the new ones. Thus we are a getting a new system of coordinates. This transformation will lead to new quasi-spin operators, i.e., R_z and $R^\pm$: $R^+ = |\bar{2}\rangle\langle\bar{1}|$, $R^- = |\bar{1}\rangle\langle\bar{2}|$, $R_z = \frac{|\bar{2}\rangle\langle\bar{2}| - |\bar{1}\rangle\langle\bar{1}|}{2}$ are describing the transitions and populations of the dressed-states $\{|\bar{2}\rangle, |\bar{1}\rangle\}$ will check the commutation relations: $\left[R^+, R^-\right] = 2R_z$ and $\left[R_z, R^\pm\right] = \pm R^\pm$, in the similar way to the bare-state basis ones. Respectively, the Hamiltonian (7) is transformed into: $H = \hbar\omega b^\dagger b + 2\hbar\bar{\Omega}R_z$, where the operator is the new generalized Rabi frequency $\bar{\Omega} = \sqrt{\frac{\bar{\Delta}^2}{4} + \Omega^2}$ whereas the bosonic annihilation operator b is expanded in power series $b = \bar{b} - i\eta S_y \sum_{k=0}^{\infty} \frac{\eta^k}{k!}\left(\bar{b}^\dagger + \bar{b}\right)^k \frac{\partial^k}{\partial\xi^k}\frac{1}{1+\xi^2}$ with bosonic operators being rotated as well, according to the unitary transformation $b^\dagger = \left[b\right]^\dagger$, $\bar{b} = UbU^{-1}$, $\bar{b}^\dagger = \left[\bar{b}\right]^\dagger$, and the small parameters defined for further derivation $\eta = \frac{g}{2\Omega}, \xi = \frac{\Delta}{2\Omega}, \bar{\Delta} = \Delta + g(b^\dagger + b)$.

In the following, we will present the cavity multiphoton quantum dynamics computed according to the master equation (6) presented above. Fig.10 presents the steady-state mean photon numbers and their second-order photon-photon correlation functions for single-photon and two-photon processes. One can notice here that these quantities are distinct from each other for single- and two-photon effects, correspondingly. In order to compare and understand the difference between single- and two-photon processes, Fig.11 presents similar effects for two- and three-photon processes, respectively. Here, it is evident that the mean-photon numbers almost overlap for the two cases under consideration, whereas their second-order correlation function is different from each other. One can proceed in the same vein with higher order photon processes.

Nevertheless, for similar considered parameters, their probabilities are small and the mean photon numbers are basically identical with ones mentioned in the description of Fig.11. Further, the photon statistics changes from super-Poissonian to quasi-thermal features, subsequently, as η in-

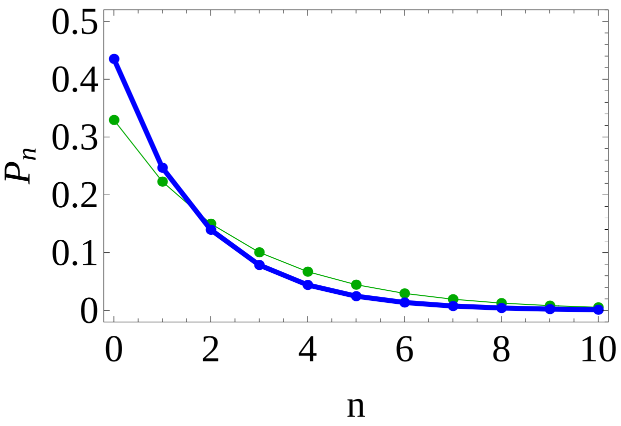


Fig. 12: The cavity photon distribution function P_n in the steady state.The green curve is plotted for $\eta = 0.09$, while the blue one for $\eta = 0.07$, respectively. Other parameters are maintained as in Fig.10, [6^a]. COLOURED ONLINE

creases with other parameters kept constant. The main conclusion drawn here is that single-, two- and three- photons processes are most feasible when other parameters are maintained constant, whereas the final cavity steady state is a quantum superposition of all those photons. Note, values different from 2 for $g^{(2)}(0)$ occur typically for higher values of η's with $\eta < 1$, ensure the creation of this final cavity state. Remark that generally the surrounding thermal mean-photon number will count linearly to the final photon flux for single-photon processes, so that an increase in the environmental temperature will lead to more output photons for the considered parameter ranges. Fig.12 displays the photon distribution function $P_n = \langle n|\bar{\rho}|n\rangle$ for the same parameters considered for the computation in Fig.10 and 11, however, for five-photon processes, i. e. $N = 5$. One can observe here that larger rations of $\eta = \frac{g}{2\Omega}$, with $\eta < 1$ lead to population of higher photon states, compare the green and the blue curves plotted for $\eta = 0.09$ and $\eta = 0.07$, respectively facilitating the generation of multiphoton states when $\frac{\kappa}{\gamma} \ll 1$. Correspondingly, P_n is small for larger n and smaller η, while $\eta < 1$, assuring convergence of the results computed by eq.(6). One can notice that the probability of a two-photon state, that is $n = 2$, is almost the same for $\eta = 0.07$ and $\eta = 0.09$, respectively, and it is higher then 0.1. One may conjecture then that a multiphoton superposition state around $n = 2$ is generated when other parameters are maintained constant. Furthermore, same results, presented in the Figs. 10, 11, 12 will be observed for moderate detunings, i.e., would not change significantly if $\xi \ll 1$.

Here, we can conclude that the presence of diagonal dipole moments, in a resonance coherently driven two-level qubit, makes achievable the coupling to the resonator mode at a completely different frequency than the input one which pumps the two-level quantum emitter, and cavity multiphoton state generation, respectively.

CONCLUSIONS AND RECOMMENDATIONS

The objectives of the thesis have been fulfilled and various new features of light-matter interaction and enhanced properties of molecular dipolar systems have been identified while studying the two-level and Λ-type three-level systems possessing a permanent non-zero dipole moment placed in quantum oscillator and interacting with external laser fields.

In correspondence with objectives stated in the introduction chapter, three different setups have been modeled in order to explain more exhaustive the impact of permanent dipole moment in all three setups and highlight the new quantum optical features of molecular dipolar systems, which were not discussed in similar researches. In the first case, one is considering a two-level system possessing permanent dipole moments and interacting with two external coherent laser fields. The first laser is near resonance with the transition frequency of the two-level system, while the second laser is close to resonance with the dressed-frequency splitting due to the first laser. In the second case, one studies the quantum dynamics of a quantum oscillator coupled with the most upper state of a three-level Λ-type system. Transitions within the three-level emitter possess orthogonal dipole moments and are coherently pumped with a single or two electromagnetic field sources, correspondingly. As a quantum oscillator in this case can serve a vibrational mode of a nanomechanical resonator embedding the three-level emitter or an electromagnetic cavity mode field if the highest energetic level of the Λ-type system incorporated in the cavity possesses a permanent dipole. In the third case, one investigates the frequency downconversion processes via a resonantly laser-pumped two-level emitter possessing non-zero permanent diagonal dipoles and is placed in a quantized microwave resonator.

The main scientific results presented in this thesis are summarized as follows:

1) The investigation of a steady-state quantum dynamics of a laser pumped two-level system possessing a non-zero permanent dipole moment involved the application of semi-classical laser-molecule dressed-state picture due to the first laser. The further dressed-state centrally symmetric transformation of the system Hamiltonian was derived. One arrived at the effective representation of the system in a frame rotating at the second laser field frequency. Using the rotating wave approximation with the respect to the second laser ω, one eliminated the vacuum modes of the electromagnetic field reservoir in the usual way by adopting the Born-Markov approximations to the studied system.

Additionally, one has plotted the resonance fluorescence spectrum of spontaneously emitted photons, squeezing spectrum and total quantum fluctuations, during the laser pumping processes of the system. New features differing from those in the case of two-level systems yet in the absence of permanent dipoles have been found. In particular, additional spectral lines are emitted and extra squeezed frequency domains are observed. The corresponding study is published in [1^a].

2) The investigation of a laser-pumped three-level Λ-type system with highest energetic level coupled with a quantum oscillator described by a single quantized leaking mode, has led ones to the identification of two distinct regimes leading to cooling and lasing effects of the quantum oscillator's degrees of freedom and have described the mechanisms determining them. Additionally, one has specified that as a quantum oscillator can serve a vibrational mode of a nanomechanical resonator containing the three-level emitter or, equivalently, an electromagnetic cavity mode field, unless the upper state of the three-level sample is embedded within the cavity, posses permanent dipole. Also it was taken into consideration, the frequency of the quantum oscillator is significantly smaller than all other frequencies involved to describe the model. Nevertheless, is of the order of the generalized Rabi frequency describing the laser-pumped three-level qubit [2^a, 3^a].

3) Following, the dressed-state picture of the three-level system, one has identified the two-resonance conditions operating the oscillator's quantum dynamics. According to the first resonant condition, the quantum oscillator's frequency is close to double generalized Rabi frequency and in the second resonant condition the qubit frequency is close to generalized Rabi frequency, respectively. For both resonant cases, one has computed the average inversion operators, the mean quanta number of the qubit and second-order correlation function analyzing the lasing and cooling phenomena occurring in the three-level system. However, one has identified the different mechanisms behind the lasing and cooling in each resonant situation [4^a].

4) We have proved that the exchange between single- or two-quanta processed followed by quantum interference effects among the induced emitter's dressed states are in charge of flexible lasing or deeper cooling effects, correspondingly. This generates also reciprocal interplay between the quantum oscillator's dynamics and the three-level emitter's quantum dynamics respectively. Additionally, if the upper state of the three-level emitter has a permanent dipole then it could couple with a single- cavity electromagnetic field mode of terahertz frequency. Another important result identified from this model is the coherent terahertz photons generation assigned as one of the possible applications resulting from this study. The first complete study on this model is published in [2^a], while the cooling regime in the three-level system was lately presented in [4^a].

5) One has investigated the quantum multiphoton dynamics of a two-level system possessing unequal permanent dipoles, placed in a leaking single-mode quantized cavity field and coupled to it. In this setup, we considered the frequencies of the interacting subsystems, namely the cavity and the emitter, are considered to belong to different frequencies range: microwave and optical domain, correspondingly, and therefore the two-level qubit couples to the resonator via its parallel dipole moments. One has demonstrated the possibility to convert photons from optical to microwave frequency domains, via resonantly pumped asymmetrical two-level quantum optical emitter placed in a quantized single-mode resonator. It was proved that cavity's multiphoton characteristics and demonstrated the feasibility for a certain output multiphoton superposition of the generated states. The present result is published in [6^a].

Considering the conlcusions above, one would propose the following recommendations:

1) The particularity of current models, consists in evaluating the impact of non-zero permanent dipole moment on the resonance fluorescence spectrum of the spontaneous emission of photons during the laser pumping processes of two-level system. Finally, one has observed the elastic photon scattering spectrum consists of three lines at $\{\omega_L, \omega_L \pm \omega\}$. The inelastic photon scattering contains up to nine spectral lines at $\omega_L, \{\omega_L \pm \omega\}, \{\omega_L \pm 2\bar{G}_R\}, \{\omega_L + \omega \pm \bar{G}_R\}, \{\omega_L - \omega \pm 2\bar{G}_R\}$. Suppression of a spectral line at the frequency of the strongly driven laser occurs due to interference effects among the induced double dressed-state transitions. Asymmetrical behaviors in the scattered light spectrum are observed as well. This is because of the population inversion in the bare state and it differs from the ordinary resonance fluorescence spectrum computed in the absence of permanent dipoles, which also modifies the squeezing spectrum for certain parameters of interest. Particularly, squeezing occurs for negative values (dark area in Fig.(3b)) and broader ranges because of the permanent dipoles. In the absence of permanent dipoles squeezing around detectors frequency ν is not observed.

2) In Λ-type three-level system possessing permanent dipole moment and embedded in an optical cavity, the frequency of the quantum oscillator is quite smaller than all other frequencies involved to describe the model; on the other hand, it is of the order of the generalized Rabi frequency identifying the laser-pumped three-level qubit. In accordance to the dressed-state base of the three-level system, we have derived two resonance conditions regulating the oscillator's quantum dynamics, specifically, when the quantum oscillator's frequency is near to the doubled generalized Rabi frequency or to the generalized Rabi frequency, correspondingly. Therefore, one recommends considering these two situations as distinct cases leading to the stationary lasing or cooling regimes for the quantum oscillator's field mode, with different mechanisms behind them.

3) If the double generalized Rabi frequency is close to the oscillator's one, then the model is in some way similar to a two-level system interacting with a quantum field mode where the spontaneous decay pumps both levels. On the other side, if the oscillator's frequency tends to the value of the generalized Rabi frequency, which is near resonance, then the sample is associated with an equidistant three-level system where the single-mode quantum oscillator interacts with both qubit's transitions. The latter case includes single- or two-quanta processes occurring simultaneously with quantum interference effects among the involved dressed states leading to more profound cooling regimes and flexible ranges for lasing effects. This is contrasting from other similar experimental schemes based on electromagnetically induced transparency processes. In this instance the model consists of an electromagnetic cavity mode, which describes the quantum oscillator, then its frequency can be in the terahertz domain and, thus, we prove an effective coherent electromagnetic field source of such photons. Thus one recommend the further extensive study of quantum interference in three-level systems.

4) It is known that lasing or cooling effects are possible within two-level system. However, the three-level system may possess an advantage as it exhibit improved features for the same parameters involved, which is a benefit when there are only certain accessible parameter ranges. Furthermore, certain realistic novel systems are explored employing the three-level model and recommended for further integration in industrial manufacturing. One recommends the developed model proper to study few coupled quantum dots and alternative systems as asymmetrical real or artificial few-level molecules possessing permanent dipoles.

5) One would suggest the further investigation of the presence of diagonal dipole moments, in a resonance coherently driven two-level qubit, because it makes achievable the coupling to the resonator mode at a completely different frequency than the input one which pumps the two-level quantum emitter, and cavity multiphoton state generation, respectively. Additionally, the proposed approach is suitable for a laser driven two-level quantum dot incorporated in an acoustical phonon cavity. In these circumstances, the manipulation of quantum states namely photons and phonons continues to be one of the main topics of modern science.

The limitation of the presented results is related to the exclusive theoretical aspect of the overall thesis, referring to already existing experimental setups.

In Chapter 2, one has presented the theoretical framework related to the dynamics of a two-level system possessing a permanent non-zero dipole moment interacting with two-laser fields. One has applied several approximations to define and explain each term of the Hamiltonian and assign it to a certain type of interaction. The main purpose was to derive the parameters of interest containing the terms responsible for the impact of permanent dipole moment. The developed approach was consequently extended in Chapters 3 and 4, where rotating wave, Born-Markov, the secular approximations were used to derive valid results plotted in corresponding graphs. Chapters

3 and 4 required certain truncation approximations due to the infinite number of quantum state, while computing the photon statistics and second order correlation function. Nevertheless, these assumptions did not affect the overall results but propose solutions to improve the parameters of existing quantum optical models.

The personal contribution of the author to presented results: The author has directly contributed to the definition of research objectives, tasks and models. She has been advised about the theoretical treatment applied to the quantum dynamics of the studied systems. She has contributed to the writting of publications drafts related to the results presented in this thesis and at various conferences.

References

[1] GLAUBER, R. J. Photon Correlations. In: *Phys. Rev. Lett.*, 1963, vol. 10, pp. 84-86.

[2] SPRING, J. B., METCALF, B. J., HUMPHREYS, P. C., KOLTHAMMER, W. S. Boson sampling on a photonic chip. In: *Science*, 2013, vol. 339, pp. 798-801.

[3] LOUDON, R., KNIGHT, P. L. Squeezed light. In: *J. Mod. Opt.*, 1987, vol. 34, pp. 709-759.

[4] ASASI, J. Enhanced sensitivity of LIGO gravitational wave detector by using squeezed state of light. In: *Nature Photonics*, 2013, vol. 7, pp. 613-619.

[5] YUEN, H., SHAPIRO, J. H. Optical communications with two-photon coherent states-part I: Quantum state propagation and quantum noise reduction. In: *IEEE Transactions on Information Theory*, 1978, vol. 24, pp. 657-668.

[6] BURNHAM, D. C., WEINBERG, D. L. Observation of simultaneity in parametric production of optical photon pairs. In: *Phys. Rev. Lett.*, 1970, vol. 25, pp. 84-87.

[7] SARGEANT, M., SCULLY, M. O., LAMB, W. E. *Laser Physics*, Addison - Wisley Pub. Co., 1974.

[8] LAKOWICZ, J. R., RAY, K., CHOWDHURY, M., SZMANCINSKI, H. Plasmon-controlled fluorescence: a new paradigm in fluorescence spectroscopy In: *The Analyst*, 2008, vol. 133, pp. 1308-1346.

[9] ACUNA, G. P., MOELLER, F. M., HOLZMEISTER, P., BEATER, S. Fluorescence enhancement at docking Sites of DNA-directed self-assembled nanoantennas. In: *Sciences*, 2012, vol. 338, pp. 506-510.

[10] LODHAL, P., MAHMOODIAN, S., STOBBE, S. Interfacing single photons and single quantum dots with photonic nanostructures. In: *Rev. Mod. Phys.*, 2015, vol. 87, pp. 347-400.

[11] HOANG, T. B., AKSELROD, G. M., MIKKELSEN, M. H. Ultrafast room-temperature single photon emission from quantum dots coupled to plasmonic nanocavities. In: *Nano. Lett.*, 2016, vol. 16, pp. 270 - 275.

[12] OKAMOTO, K., NIKI, I., SHVARTSER, A., NARUKAWA, Y., MUKAI, T. Surface-plasmon-enhanced light emitters based on InGaN quantum wells. In: *Nat. Mater.*, 2004, vol. 3, pp. 601-605.

[13] KREINBERG, S., GRBEŠIĆ, T., STRAUß, M. Quantum-optical spectroscopy of a two-level system using an electrically driven micropillar laser as a resonant excitation source. In: *Light Sci. Appl.*, 2018, vol. 41, pp. 1-9.

[14] HE, Y.-L. Multiphoton Rabi oscillations between highly excited Stark states of potassium. In: *Phys. Rev. A.*, 2011, vol. 84, pp. 053414/ 1-7.

[15] TAMINIAU, T. H., KARAVELI, S.; VAN HULST, N. F., ZIA, R. Quantifying the magnetic nature of light emission. In: *Nat. Commun.*, 2012, vol. 3, pp. 9791/1-6.

[16] AIGOUY, L., CAZE, A., GREDIN, P., MORTIER, M., CARMINATI, R. Mapping and quantifying electric and magnetic dipole luminescence at the nanoscale. In: *Phys. Rev. Lett.*, 2014, vol. 113, pp. 0761101/1-5.

[17] CALDERON, O. G., GUTIERREZ-CASTREJON, R.; GUERRA, J. M. High harmonic generation induced by permanent dipole moment. In: *IEEE J. Quantum Electron.*, 1999, vol.35, pp. 47-52.

[18] KONDO, A. E., BLOKKER, V. M., MEATH, W. J. Permanent dipole moments and two-color multiphoton resonances in the two-level molecule: the rotating wave approximations versus exact results. In: *J. Chem. Phys.*, 1992, vol. 96, pp. 2544-2555.

[19] BESSEGA, M. C., PAZ, J. L., HERNANDEZ, A. J., CARDENS, A. E. Study of additional resonances in frequency space and permanent dipole effects on non-degenerate four-wave mixing signals. In: *Phys. Lett. A.*, 1995, vol. 206, pp. 305-310.

[20] HOERNER, C., LAVINE, J. P.; VILLAYES, A. A. Theoretical description of two-photon phase conjugation in polar molecules. In: *Phys. Rev. A*, 1993, vol. 48, pp. 1564-1572.

[21] SAVENKO, I. G., KIBIS, O. V., SHLEYKH, I. A. Assymetric quantum dot in a microcavity as a nonlinear optical element. In: *Phys. Rev. A*, 2012, vol. 85, pp. 053815/1-6.

[22] AVETISSIAN, H. K., MKRTCHIAN, G. F., POGOSYAN, M. G. Creation of coherent superposition states in hydrogenlike ions via multiphoton resonant photonic field. In:*Phys. Rev. A.*, 2006, vol. 73, pp. 063413/1-7.

[23] AVETISSIAN, H. K., MKRTCHIAN, G. F. Two-level system with broken inversion symmetry coupled to a quantum harmonic oscillator. In: *Phys. Rev. A.*, 2013, vol. 88, pp. 043811/1-6.

[24] MACOVEI, M., MISHRA, M., KEITEL, C.H. Population inversion in two-level systems possessing permanent dipoles. In: *Phys. Rev. A.*, 2015, vol. 92, pp. 013846/1-5.

[25] BAVLI, R., BAND, Y. B. Nonlinear absorption and dispersion in a two-level system with permanent dipole moments. In: *Phys. Rev. A.*, 1991, vol. 43, pp. 5039-5043.

[26] ANTON, M. A., MAEDE-RAZAVI, S., CARRENO, F., THANOPOULOS, I., PASPALAKIS, E. Optical and microwave control of resonance fluorescence and squeezing spectra in a polar molecule. In: *Phys. Rev. A.*, 2017, vol. 96, pp. 063812/1-16.

[27] BROWN, A., MEATH, W. J. Two-color pulsed-laser phase control in dipolar molecules: Rotating-wave approximation versus exact results. In: *Phys. Rev. A.*, 2002, vol. 65, pp. 063401/ 1-12.

[28] KOCINAC, S., IKONIC, Z., MILANOVIC, V. Second harmonic generation at the quantum-interference induced transparency in semiconductor quantum wells: the influence of permanent dipole moments. In: *IEEE Journal of Quantum Electronics*, 2001, vol. 37, pp. 873-876.

[29] LEE, M., WANKE, M. Searching for a solid-state terahertz technology. In: *Science*, 2007, vol. 64, pp. 64-65.

[30] LAWANDE, Q. V., LAWANDE, S. V. Squeezing in Cooperative Resonance Fluorescence. In:*Phys. Rev. A*, 1988, vol.38, pp. 800-807.

[31] AGARWAL, G.S. *Quantum Optics*, Cambridge University, 2014.

[32] WALLS, D.F., MILBURN, G.J. *Quantum Optics*, Springer, 2008.

[33] ZAKREZEWSKI, J., LEWENSTEIN, M., MOSSBERG, T.W. Theory of dressed-state laser. I. Effective Hamiltonians and stability properties. In: *Phys. Rev. A*, 1991, vol.44, pp. 7717-7731.

LIST OF AUTHOR’S PUBLICATIONS

Publications in Scientific Journals:

[1^{a}] MIRZAC, A., CIORNEA, V., MACOVEI, M. Non-classical light scattered by laser-pumped molecules possessing permanent dipoles.
In: *Moldavian Journal of Physical Sciences*, 2018, vol. 1-2(17), pp. 95-104. ISSN 1810-648X /ISSNe 2537-6365

[2^{a}] MIRZAC, A., MACOVEI, M. Dynamics of a quantum oscillator coupled with a three-level Λ-type emitter. In: *J. Opt. Soc. Am. B*, 2019, vol. 9(36), pp. 2473-2480.
ISSN 0740-3224 (IF: 2,180).

[3^{a}] MÎRZAC, A. Dinamica răcirii cuantice a unui oscilator cuplat cu un atom artificial. In:
Revista de Știință, Inovare Cultură și Artă ”Akademos”, 2019, vol. 4(55), pp. 16 - 19.
ISSN 1857-0461.

[4^{a}] MIRZAC, A., MACOVEI, M. Steady-state behaviors of a quantum oscillator coupled with a three-level emitter. In: *Tiginyanu I., Sontea V., Railean S. (eds) 4th International Conference on Nanotechnologies and Biomedical Engineering. ICNBME 2019. IFMBE Proceedings, Springer, Cham.*, 2020, vol. 77, pp.677-680. ISSN 1433-9277.

[5^{a}] MIRZAC, A., MIHAESCU, T., MACOVEI, M., ISAR, A. Interferometric power of gaussian systems in a squeezed thermal bath. In: *Romanian Journal of Physics*, 2020, vol. 1-2(65), pp. 95-104. ISSN 1221-146X.

[6^{a}] MIRZAC, A., CÂRLIG, S., MACOVEI, M. Microwave multiphoton conversion via coherently driven permanent dipole systems. In: *Phys.Rev.A*, 2021, vol.103, pp.043719/1-7.

Conference Proceedings and Abstracts:

[7^{a}] MIRZAC, A. Kinetics of molecular systems with permanent dipole moment. In: *Tendințe contemporane ale dezvoltării științei: viziuni ale tinerilor cercetători. Ediția 5*, Chișinău, Republica Moldova, 2016, vol.2, pp. 46-48.

[9^{a}] MIRZAC, A., MACOVEI, M. Resonance fluorescence spectrum for a pumped dipolar molecule. In: *Materials Science and Condensed Matter Physics, Ediția a 8 - a*, Chișinău, Republica Moldova, 2016, pp.62. ISBN 978-9975-9787-1-2.

[10^{a}] MÎRZAC, A., MACOVEI, M. Squeezing in fluorescence spectrum of pumped molecular systems with permanent dipole. In: *Humbold Kolleg Multidisciplinarity in Modern Science*

for the Benefit of Society, Chișinău, Republica Moldova, 2017, p.58.
ISBN 978-9975-9787-1-2.

[11^a] MÎRZAC, A. Resonance fluorescence spectrum in pumped molecular systems with permanent dipole. In: *Tendințe contemporane ale dezvoltării științei: viziuni ale tinerilor cercetători, Ediția 6*, Chișinău, Republica Moldova, 2017, pp. 79-84.

[12^a] MÎRZAC, A., CIORNEA, V., MACOVEI, M. Quantum light features scattered by pumped two-level systems with permanent dipoles. In: *Telecommunications, Electronics and Informatics, Ediția 6*, Chișinău, Republica Moldova, 2018, pp. 89-91.
ISBN 978-9975-45-540-4

[13^a] MÎRZAC, A. Quantum fluctuations in laser pumped molecular systems with permanent dipole. In: *Tendințe contemporane ale dezvoltării științei: viziuni ale tinerilor cercetători, Ediția 7*, Chișinău, Republica Moldova, 2018, vol.1, pp. 59-64. ISBN 978-9975-108-45-4

[14^a] MÎRZAC, A., MACOVEI, M. Non-classical light features scattered by laser pumped molecules with permanent dipoles. In: *Materials Science and Condensed Matter Physics, Ediția a 9-a*, Chișinău, Republica Moldova, 2018, p. 64

[15^a] MÎRZAC, A. Steady-state dynamics of a quantum oscillators coupled with a three-level emitter. In: *Conferinţa tehnico-ştiinţifică a studenţilor, masteranzilor şi doctoranzilor*, Chișinău, Republica Moldova, 2019, vol. 1, p. 378. ISBN 978-9975-45-588-6

[16^a] MÎRZAC, A., Macovei, M. Lasing and cooling effects of a quantum oscillator coupled with a three-level Λ – type system. In: *International Conference on Advanced Scientific Computing*, Sinaia, Romania, 2019, p. 59 ISBN 978-973-0-30119-9.

SUMMARY

to the thesis ”Quantum dynamics in molecular dipolar systems”, presented by Alexandra Mîrzac for conferring the scientific degree of Ph.D. in Physics, Speciality 131.01 ”Mathematical Physics”, Chişinau, 2021.

The thesis has been written in English language and consists of the introduction, 4 chapters, general conclusions and recommendations, and the list of 205 references. The thesis contains 134 pages of basic text, 19 figures and 141 formulae. The results presented in the thesis are published in 16 scientific papers.

Key words: two-level system, three-level Λ-system, permanent dipole moment, resonance, fluorescence, squeezing, terahertz lasing, multi-quanta processes, quantum interference, multiphoton conversion, quantum emitter, super-Poissonian statistics.

The goal: The detection of new quantum dynamical properties in two and three-level Λ-type systems possessing a non-zero permanent dipole moment strongly coupled with quantum optical cavity or opto-mechanical resonators.

Research objectives: The calculation of squeezing effects in the resonance fluorescence processes of laser-pumped two-level system possessing a permanent dipole moment; The determination of the total quantum fluctuation spectra of laser-pumped dipolar two-level systems; The investigation of a laser-pumped three-level Λ-type system having the upper state coupled with a quantum oscillator described by a single quantized leaking mode; The identification of three-level model particularities leading to lasing and cooling effects; The demonstration of quantum interference effects induced by emitter's dressed states responsible for flexible lasing and deeper cooling effects; The investigation of frequency conversion from optical to microwave region, via the resonant pumping of an asymmetrical two-level system incorporated in a quantized single-mode resonator; The demonstration of multiphoton features of cavity quantum dynamics containing an asymmetric two-level system using certain multiphoton superposition of generated states.

Scientific novelty and originality of the results: the new features of resonance fluorescence spectrum of spontaneously emitted photons by dipolar two-level system were demonstrated; two distinct mechanisms of lasing and cooling based on single- or two-quanta processes where detected in the three-level Λ-type system; conversion of photons from optical to microwave domains, via resonantly pumped asymmetrical two-level quantum emitter embedded in a quantized single-mode resonator.

The main scientific problem solved consists in computing and analyzing the quantum dynamical properties of few level atomic systems possessing a permanent dipole moment interacting with external coherent laser field.

Theoretical significance and applicative value: in the thesis, one has investigated the steady state-quantum dynamics of a laser pumped two-level system possessing a non-zero permanent dipole moment. New features of the dipolar two-level system have been found in the resonance fluorescence spectrum, squeezing spectrum and total quantum fluctuations.

The model of a laser-pumped three-level Λ-type system with highest energetic level coupled with a quantum oscillator described by a single quantized leaking mode has been investigated. Two distinct regimes leading to cooling and lasing effects of the model have been identified. In the first regime, the model functions as a two-level system. Whereas in the second regime, the model evolves into a three-level equidistant system.

The quantum multiphoton dynamics of a two-level system possessing unequal permanent dipoles, placed in a leaking single-mode quantized cavity field and coupled to it has been investigated. The photons conversion from optical to microwave frequency domains was proved.

The implementation of the scientific results: the research presented in this thesis have been successfully implemented in the framework of the national project (15.817.02.09F) also with support of Moldavian National Agency for Research and Development, grant No. 20.80009.5007.07 and National Scholarship of World Federation of Scientists in Moldova.

ADNOTARE

la teza ”Studiul dinamicii cuantice în sistemele moleculare dipolare”, elaborată de Alexandra Mîrzac pentru conferirea gradului științific de doctor în științe fizice la specialitatea 131.01 ”Fizică matematică”, Chișinău, 2021.

Teza este scrisă în limba engleză și constă din introducere, 4 capitole, concluzii generale și recomandări, și lista a 205 referințe bibliografice. Teza conține 134 pagini de text de bază, 19 figuri și 141 formule. Rezultatele prezentate în teză sunt publicate în 16 lucrări științifice.

Cuvinte cheie: sistem cu două nivele, Λ-sistem cu trei niveluri, dipol permanent, fluorescența la rezonanță, comprimare, laser terahertz, procese cuantice multiple, interferență cuantică, conversia multifotonică, emițător cuantic, statistică super-Poissoniană.

Scopul tezei: Detectarea proprietăților noi în dinamica cuantică a sistemelor cu două niveluri și trei niveluri energetice de tip Λ care posedă dipol permanent nenul și sunt cuplate cu cavitatea optică cuantică sau rezonator opto-mecanic.

Obiectivele tezei: Demonstrarea efectelor de comprimare în spectrul fluorescenței de rezonanță a sistemelor cu două niveluri pompate laser; Determinarea spectrului fluctuațiilor cuantice totale a sistemelor dipolare cu două niveluri; Identificarea mecanismelor de emisie laser și răcire în domeniul THz în sistem cu trei nivele energetice de tip Λ cuplat prin dipol permanent nenul cu un oscilator cuantic; Demonstrarea efectelor de interferență cuantică care induc emisie laser și răcire cuantică într-un domeniu extins de frecvențe; Cercetarea metodei de conversie optică de la domeniul optic spre domeniul microundelor, prin pomparea rezonantă a sistemlor asimetrice cu două nivele încorporat de un rezonator cuantic unimod; Demonstrarea proprietăților multifotonice ale cavității cuantice care conține un sistem asimetric cu două niveluri, prin suprapunerea multifotonică a stărilor generate.

Noutatea științifică și originalitatea rezultatelor: au fost demonstrate proprietățile noi ale spectrului fluorescenței de rezonanță al fotonilor emiși spontan de către un sistem dipolar cu două niveluri; au fost determinate două mecanisme distincte ale emisiei laser și răcirii cuantice într-un sistem de tip Λ cu trei niveluri cu dipol permanent nenul, implicând procese cuantice unitare și binare; a fost demonstrată conversia frecvenței fotonilor din domeniul optic în domeniul microundelor prin pomparea rezonantă a unui emițător asimetric cu două niveluri incorporat într-un rezonator cuantic unimodal.

Problema științifică soluționată constă în calculul și analiza proprietăților dinamicii cuantice a sistemelor cu două și trei niveluri energetice, care posedă dipol permanent nenul și interacționează cu câmpuri externe coerente laser.

Semnificația teoretică și valoarea aplicativă: în această teză, este investigată dinamica complexă cuantică a unui sistem cu două niveluri, cu dipol permanent nenul, interacționând cu câmp laser. Au fost determinate noi proprietăți ale sistemului dipolar cu două niveluri prin observarea unor aspecte distincte în spectrele fluorescenței la rezonanță ale fotonilor emiși spontan, comprimării fluorescenței de rezonanță și fluctuațiilor cuantice totale, față de cazul neglijării dipolului permanent. A fost cercetat modelul unui sistem de tip Λ cu trei niveluri energetice cu nivel superior cuplat cu un oscilator cuantic unimodal. În cadrul acestui model au fost identificate două cazuri distincte de emisie laser și de răcire laser în domeniul THz. În primul caz, modelul este redus la un sistem cu două nivele. În al doilea caz, modelul este extins la un sistem echidistant de trei niveluri, în care frecvența qubitului este apropiată de frecvența generalizată Rabi. A fost modelată dinamica multifotonică a sistemului dipolar cu două niveluri plasat într-o cavitate optică cuantică și cuplată cu aceasta prin dipol permanent. A fost demonstrată modularea frecvenței fotonilor din domeniul optic în domeniul microundelor prin pomparea rezonantă a emițătorului optic asimetric cu două niveluri plasat într-un rezonator cuantic unimodal.

Implementarea rezultatelor științifice: studiile prezentate în această teză au fost implementate cu succes în cadrul proiectului național (15.817.02.09F), cu suportul financiar al Agenției Naționale pentru Cercetare şi Dezvoltare, grant Nr.20.80009.5007.07 și cu suportul Bursei Naționale oferită de Federația Mondială a Savanților în Moldova.

АННОТАЦИЯ

к диссертации «Исследования квантовой динамики молекулярных диполярных систем», представленной Александрой Мырзак на соискание ученой степени доктора физических наук по специальности 131.01 «Математическая физика», Кишинэу, 2021.

Диссертация написана на английском языке и состоит из введения, четыреёх глав, общих заключений и рекомендаций, и списка цитируемой литературы из 205 источников. Диссертация содержит 134 страниц основного текста, 19 графиков и 141 формул. Результаты диссертационной работы опубликованы в 16 научных публикациях.

Ключевые слова: двухуровневая система, трёхуровневая система Λ-типа, постоянный диполь, резонантная флюресценция, сжатие, терагерцовое лазерное излучение, многоквантовые процессы, квантовая интерференция, генерация фотонов, генерация фононов, многофотонное преобразование, квантовый излучатель, одномодовый резонатор, многофононная генерация, супер-Пуассоновская статистика, квази-Пуассоновская статистика.

Цель диссертации: Обнаружение новых квантовых динамических свойств в двух- и трехуровневых системах Λ-типа, обладающих ненулевым постоянным дипольным моментом, сильно связанным с квантовым оптическим резонатором или оптико-механическими резонаторами.

Задачи диссертации: Расчет и моделирование эффектов сжатия в процессах резонансной флуоресценции двухуровневой системы с лазерной накачкой, обладающей постоянным дипольным моментом; Моделирование спектров полной квантовой флуктуации в дипольных двухуровневых систем с лазерной накачкой; Исследование новых свойств трехуровневой системы Λ-типа с лазерной накачкой приводящих к гибкому лазерному излучению и охлаждению; Доказательство квантово-интерференционных эффектов ведущих к гибкому лазерному излучению и более глубокому лазерному охлаждению; Исследование преобразования частоты из оптической области в микроволновую с помощью резонансной накачки несимметричной двухуровневой системы; Доказательство многофотонных особенностей квантовой динамики резонатора, содержащего несимметричную двухуровневую систему, с использованием некоторой многофотонной суперпозиции генерируемых состояний.

Научная новизна и оригинальность результатов: доказанны новые особенности спектра резонансной флуоресценции спонтанно испускаемых фотонов диполярной двухуровневой системой; в трехуровневой системе Λ-типа обнаружены два различных механизма лазерного излучени и охлаждения, включающие одно- или двухквантовые процессы; доказанно преобразование фотонов из оптической в микроволновую область с помощью асимметричного двухуровневого квантового излучателя с резонансной накачкой, встроенного в квантованный одномодовый резонатор.

Основная научная задача, решаемая диссертацией, состоит в вычислении и анализе квантовых динамических свойств малоуровневых атомных систем, обладающих постоянным дипольным моментом, взаимодействующими с внешним когерентным лазерным полем.

Теоретическая значимость и прикладная ценность: в диссертации исследовались новые особенности спектров резонансной флуоресценции спонтанно испускаемых фотонов, сжатия и полных квантовых флуктуаций в двухуровневой системе с лазерной накачкой, обладающей ненулевым постоянным дипольным моментом

Исследованы два различных механизма лазерного излучения и охлаждения в трехуровневой системе Λ-типа с лазерной накачкой. Согласно им, модель обладает одновременно свойствами двухуровневой и трёхуровневой эквидистантной системы.

Исследована квантовая многофотонная динамика двухуровневой системы с неодинаковыми постоянными диполями и преобразование фотонов из оптической в микроволновую частотную область с помощью асимметричного двухуровневого квантово-оптического излучателя с резонансной накачкой, помещенного в квантованный одномодовый резонатор.

Внедрение научных результатов: исследования, представленные в этой диссертации, были успешно внедренны в рамках национального проекта (15.817.02.09F), а также при поддержке Нац. Агентства по Исследованиям и Развитию Молдовы, грант (20.80009.5007.07) и Нац. Стипендии Всемирной Федерации Ученых (Швейцария) в Молдове.

QUANTUM DYNAMICS IN MOLECULAR DIPOLAR SYSTEMS

MÎRZAC ALEXANDRA

131.01 - MATHEMATICAL PHYSICS

Summary of doctoral thesis in physics

Approved for printing: 27.04.2022
Paper Size 60x84 1/16
Offset paper. Offset typing.
Circulation: 30 copies
Print Sheets 2,0.
Order no. 35

Tipografia MS Logo,
str. Columna 145, Chişinău