Quantum light features scattered by pumped two-level systems with permanent dipoles.

A. Mirzac[1], V. Ciornea[1], M. A. Macovei[1]
[1]Institute of Applied Physics, Academiei str. 5, MD – 2028, Chișinău, Moldova
mirzacalexandra71@phys.asm.md

Abstract **— Here we investigate the quantum fluctuations in the light scattered by laser driving two – level systems with permanent dipoles. Particularly, we have observed additional quantum features due to existence of the permanent dipoles.**

Key words **— two-level system, squeezing, fluctuations, quantum dot, resonance fluorescence, permanent dipole**

I. Introduction

Researchers from the quantum optics community have attracted great attention, during the last five decades, to the resonance fluorescence which is conceptually simple and leading to a wide range of intriguing phenomena like photon anti-bunching, squeezing or various other effects in the Mollow-triplet emission [1,2]. Additionally, the potential applications in quantum computing of systems such as semiconductor quantum dots, as wel as trapped atoms and ions, has renewed the interest on this topic recently.

The prediction of the fluorescence spectrum of a single atom under coherent excitation, resulting in canonical phenomena such as Mollow triplet, represents a fundamental success of quantum community. Since, the Mollow triplet provides a clear picture of coherent light-matter coupling, in recent years such spectra have been widely applied to detect quantum coherence in artificial atoms based on quantum dots and superconducting qubits. The main nonclassical feature of the resonance fluorescence spectrum is the squeezing of the field quadratures of a two-level system, which was theoretically proposed by Walls and Zoller and later experimentally proved [3]. Graviational wave detection and quantum computing are examples of its feasable applications. The squeezing of the fluorescent field has been widely studied in two – and three – level atoms driven by laser. Thus, taking into account the fast developement of quantum informatics, squeezed states of the radiation field have been recongized as tools for quantum information processing.

II. Theoretical Model

We are considering a two-level system with permanent dipole moments, which is 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 dressed-frequency splitting due to the first laser respectively. The analytical formalism applies equally to spin, asymmetric semiconductor quantum dot or other alternative systems promising wider applications. The Hamiltonian describing such a model, in the frame of rotating wave approximation at the first laser frequency ω_L, as well in the dipole approximation is [4]:

$$H = \sum_k \hbar(\omega_k - \omega_L) a_k^\dagger a_k + \hbar\Delta S_z + \hbar\Omega(S^+ - S^-) + \hbar G S_z \cos(\omega_2 t) + i\sum_k(\overrightarrow{g_k}\cdot\vec{d})(a_k^\dagger S^- - a_k S^+) \quad (1)$$

This Hamiltonian consists of three components, which are: the free energies of the environmental electromagnetic vacuum modes and molecular subsystems together with laser-molecule interaction Hamiltonian. Here, $\Omega = dE_1/2\hbar$ 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 fourth term accounts for the second laser interacting at the frequency ω and amplitude E_2 with the molecular system due to the presence of permanent dipoles incorporated in $G = (d_{22} - d_{11})E_2/\hbar$. The last term pictures the interaction of the molecular subsystem with the environmental vacuum modes of the electromagnetic field [4-10]. Further on, vector $\vec{g} = \sqrt{2\pi\hbar\omega_k/V}\overrightarrow{e_\lambda}$ is the molecule-vacuum strength, where $\overrightarrow{e_\lambda}$ is considered the photon polarization vector and $\lambda \in \{1,2\}$, whereas V is the quantization volume. $\Delta = \omega_{21} - \omega_L$ is the laser filed detuning from the molecular transition frequency ω_{21}. We are using the following bare state operators $S^+ = |2\rangle\langle 1|$ and $S^- = [S^+]^\dagger$ are verifying the commutation relations $[S^+, S^-] = 2S_z$ and$[S_z, S^\pm] = \pm S^\pm$. The bare-state inversion operator is given by the following relation $S_z = (|2\rangle\langle 2| - |1\rangle\langle 1|)/2$. The S^-operator brings an atom in the upper state into the lower state, whereas S_+ brings an atom in the lower state into the upper state.

The interaction energy in equation (1) consisted of four terms, is obtained in the rotating wave approximation. The $a_k^\dagger S^-$ term describes the process in which the atom is taken from the upper state into lower and a photon in mode $\boldsymbol{k}$ is created. The term $a_k S^+$ describes the opposite process. Terms which were dropped according to rotating wave approximation, are: $a_k S^-$ and $a_k^\dagger S^+$. $a_k S^-$ describes the process in which the atom makes a transition from the upper to the lower level, annihilating a photon, which results in the loss of approximately $2\hbar\omega$ energy. In the same way, the term $a_k^\dagger S^+$ result in the gain of $2\hbar\omega$ energy. These nonconserving energy terms were dropped according to rotating wave approximation. The excited and ground state of the molecule are $|2\rangle$ and $|1\rangle$, while $a_k^\dagger$ and

a_k are the creation and the annihilation operator of the k_{th} electromagnetic field mode, and are satisfying the standard bosonic commutation relations, exactly $[a_k, a^\dagger_{k'}] = \delta_{kk'}$, and $[a_k, a_{k'}] = [a^\dagger_k, a^\dagger_{k'}] = 0$.

Taking into account the complexity of the proposed problem, we have performed two transformation of initial Hamiltonian within Rotating Wave and Born-Markov approximations. We make the first transformation in "single dressed" state basis of the excited and fundamental molecule state, due to the pumping of the first laser:

$$|1\rangle = \sqrt{\frac{1}{2}\left(1+\frac{\Delta}{2\bar{\Omega}}\right)}\,|\bar{1}\rangle + \sqrt{\frac{1}{2}\left(1-\frac{\Delta}{2\bar{\Omega}}\right)}\,|\bar{2}\rangle, \qquad (2)$$

$$|2\rangle = \sqrt{\frac{1}{2}\left(1-\frac{\Delta}{2\bar{\Omega}}\right)}\,|\bar{1}\rangle + \sqrt{\frac{1}{2}\left(1+\frac{\Delta}{2\bar{\Omega}}\right)}\,|\bar{2}\rangle, \qquad (3)$$

where the new Rabi frequency is: $\bar{\Omega} = \sqrt{\Omega_1^2 + (\Delta/2)^2}$.
The second transformation in "double dressed' state base is:

$$|\bar{1}\rangle = \sqrt{\frac{1}{2}\left(1+\frac{\bar{\Delta}}{\bar{G}_R}\right)}\,|\tilde{1}\rangle + \sqrt{\frac{1}{2}\left(1-\sqrt{1-\frac{\bar{G}}{\bar{G}_R}}\right)}\,|\tilde{2}\rangle, \qquad (4)$$

$$|\bar{2}\rangle = \sqrt{\frac{1}{2}\left(1-\sqrt{1-\frac{\bar{G}}{\bar{G}_R}}\right)}\,|\tilde{1}\rangle + \sqrt{\frac{1}{2}\left(1+\frac{\bar{\Delta}}{\bar{G}_R}\right)}\,|\tilde{2}\rangle, \qquad (5)$$

and we obtain the following Master Equation [4]:

$$\frac{d}{dt}\langle Q(t)\rangle = i\bar{G}_R\langle[\tilde{R}_z, Q]\rangle$$
$$-\bar{\Gamma}_0\{\langle\tilde{R}_z[\tilde{R}_z, Q]\rangle + \langle[Q,\tilde{R}_z]\tilde{R}_z\rangle\} \qquad (6)$$
$$-\bar{\Gamma}_+\{\langle\tilde{R}^+[\tilde{R}^-, Q]\rangle + \langle[Q,\tilde{R}^+]\tilde{R}^-\rangle\}$$
$$-\bar{\Gamma}_-\{\langle\tilde{R}^-[\tilde{R}^+, Q]\rangle + \langle[Q,\tilde{R}^-]\tilde{R}^+\rangle\}$$

where $\gamma = \frac{1}{2}\frac{4d^2\omega_0^3}{3\hbar c^3}$; $\bar{G}_R = \sqrt{\bar{\Delta}^2 + \bar{G}^2}$; $\bar{\Delta} = \bar{\Omega} - \omega/2$ and dropping off the rapid oscillating terms: $e^{\pm i\omega t}$, $e^{\pm 2i\bar{G}_R t}$, $e^{\pm 2i\omega t}$, $e^{\pm i(2\bar{G}_R \pm\omega)t}$, $e^{\pm i(2\bar{G}_R\pm 2\omega)t}$, $e^{\pm i(4\bar{G}_R\pm 2\omega)t}$, $e^{\pm i(4\bar{G}_R\pm\omega)t}$, $e^{\pm 4i\bar{G}_R t}$, we get the analytical expressions for the spontaneous emission rates in "double-dressed" state base:

$$\bar{\Gamma}_0 = \frac{1}{4}\gamma\omega_L sin^2 2\theta cos^2 2\bar{\theta} + \frac{1}{4}\gamma(\omega_L+\omega)sin^2 2\bar{\theta} cos^4\theta + \frac{1}{4}\gamma(\omega_L-\omega)sin^2 2\bar{\theta} sin^4\theta, \qquad (7)$$

$$\bar{\Gamma}_+ = \frac{1}{4}\gamma(\omega_L+2\bar{G}_R)sin^2 2\bar{\theta} sin^2 2\theta + \frac{1}{4}\gamma(\omega_L+\omega+2\bar{G}_R)cos^4\bar{\theta}cos^4\theta + \frac{1}{4}\gamma(\omega_L-\omega+2\bar{G}_R)sin^4\theta sin^4\bar{\theta}, \qquad (8)$$

$$\bar{\Gamma}_- = \frac{1}{4}\gamma(\omega_L-2\bar{G}_R)sin^2 2\theta sin^2 2\bar{\theta} + \frac{1}{4}\gamma(\omega_L+\omega-2\bar{G}_R)cos^4\theta sin^4\bar{\theta} + \frac{1}{4}\gamma(\omega_L-\omega-2\bar{G}_R)sin^4\theta cos^4\bar{\theta}, \qquad (9)$$

where $\tan 2\theta = \frac{2\bar{\Omega}}{\Delta}$ and $\cot 2\bar{\theta} = \frac{\bar{\Delta}}{\bar{G}}$.

In the next section, we shall calculate the squeezing effects in the resonance fluorescence spectrum based on Master Equation (6). Notice that the resonance fluorescence spectrum may contain up to nine lines incoherently scattered by the laser pumped molecules as well as three coherent scattered ones [4].

III. Results and Discussions.

The steady – state spectrum of squeezing is then defined as [11]:

$$S_\varphi(\nu) = (\gamma/|\mu|^2)\int_0^\infty [exp(i\nu\tau) + exp(-i\nu\tau)] \times \lim_{t\to\infty}\Gamma_\varphi(t+\tau, t)\,d\tau \qquad (10)$$

where $\Gamma_\varphi(t+\tau,t) = \langle :\Delta E_\varphi(t+\tau)\Delta E_\varphi(t):\rangle$.
Reduced quantum fluctuations are phase dependent, which is expressed by the weak oscillating terms of the field emitted to the detector:

$$E_\varphi(t) = \frac{E^{(+)}(t)exp(i\varphi) + E^{(-)}(t)exp(-i\varphi)}{2} \qquad (11)$$

φ is the phase angle and $E^{(\pm)}$ are the negative and positive amplitudes of the field. Particularly we compute:

$$\Gamma_\varphi(t+\tau,t) = \frac{1}{4}\langle:\left(E^{(+)}(t+\tau)e^{i\varphi} - E^{(-)}(t+\tau)e^{-i\varphi} - \langle E^{(+)}(t+\tau)\rangle e^{i\varphi} - \langle E^{(-)}(t+\tau)\rangle e^{-i\varphi}\right) \times \left(E^{(+)}(t)e^{i\varphi} - E^{(-)}(t)e^{-i\varphi} - \langle E^{(+)}(t)\rangle e^{i\varphi} - \langle E^{(-)}(t)\rangle e^{-i\varphi}\right):\rangle \qquad (12)$$

Taking into account that: $E^+ \sim a \sim S^-$; $E \sim a^+ \sim S^+$, the radiated field $E^{(+)}(t)$ can be substituted by $\mu S_-(t)$, where μ is a geometric factor. So, in this way, we obtain [11]:

$$\Gamma_\varphi(t+\tau,t) = \frac{|\mu|^2}{4}\{e^{2i\varphi}(\langle S^-(t+\tau)S^-(t)\rangle - \langle S^-(t+\tau)\rangle\langle S^-(t)\rangle) +$$
$$+e^{-2i\varphi}(\langle S^+(t)S^+(t+\tau)\rangle - \langle S^+(t)\rangle\langle S^+(t+\tau)\rangle) +$$
$$+(\langle S^+(t+\tau)S^-(t)\rangle - \langle S^+(t+\tau)\rangle\langle S^-(t)\rangle) +$$
$$+(\langle S^+(t)S^-(t+\tau)\rangle - \langle S^+(t)\rangle\langle S^-(t+\tau)\rangle)\} \qquad (13)$$

Using (2)-(5) and the solutions of the Master equations (6):

$$\langle\tilde{R}^-(\tau)\tilde{R}^+\rangle = \frac{1}{2}\left(1-\langle\tilde{R}_z\rangle_s\right)e^{-(2i\bar{G}_R+\bar{\Gamma}_s)\tau}, \qquad (14)$$

$$\langle\tilde{R}^+\tilde{R}^-(\tau)\rangle = \frac{1}{2}\left(1+\langle\tilde{R}_z\rangle_s\right)e^{-(2i\bar{G}_R+\bar{\Gamma}_s)\tau}, \qquad (15)$$

$$\langle\tilde{R}^+(\tau)\tilde{R}^-\rangle = \frac{1}{2}\left(1+\langle\tilde{R}_z\rangle_s\right)e^{(2i\bar{G}_R-\bar{\Gamma}_s)\tau}, \qquad (16)$$

$$\langle\tilde{R}^-\tilde{R}^+(\tau)\rangle = \frac{1}{2}\left(1-\langle\tilde{R}_z\rangle_s\right)e^{(2i\bar{G}_R-\bar{\Gamma}_s)\tau}, \qquad (17)$$

we arrive at the analytical expression for the spectrum of squeezing:

$$S_\varphi(\nu) = \frac{\gamma}{4}\times$$
$$\{(\langle\tilde{R}_z^2\rangle_s - \langle\tilde{R}_z\rangle_s^2)[\sin^2 2\theta\cos^2 2\bar{\theta}(1+\cos 2\varphi)\chi_1(\nu)$$
$$+\frac{1}{2}\sin^2 2\bar{\theta}\left(\cos^4\theta + \sin^4\theta - \frac{1}{2}\sin^2 2\theta\cos 2\varphi\right)\chi_2(\nu) +$$
$$+\frac{1}{2}\sin^2 2\theta\sin^2 2\bar{\theta}(1+\cos 2\varphi)\chi_3(\nu) +$$
$$+\sin^4\bar{\theta}\left(\cos^4\theta + \sin^4\theta - \langle\tilde{R}_z\rangle_s\cos 2\theta - \frac{1}{2}\sin^2 2\theta\cos 2\varphi\right)\chi_4(\nu) +$$
$$+\cos^4\bar{\theta}\left(\cos^4\theta + \sin^4\theta + \langle\tilde{R}_z\rangle_s\cos 2\theta - \frac{1}{2}\sin^2 2\theta\cos 2\varphi\right)\chi_5(\nu) -$$
$$-\frac{1}{2}\sin^2 2\theta\sin^2 2\bar{\theta}\sin 2\varphi\,\langle\tilde{R}_z\rangle_s\chi_6(\nu) +$$
$$-\frac{1}{2}\sin^2 2\theta\sin^4\bar{\theta}\sin 2\varphi\,\langle\tilde{R}_z\rangle_s\chi_7(\nu) +$$

$$+\frac{1}{2}\sin^2 2\,\theta\cos^4\bar{\theta}\sin 2\,\varphi\,\langle\tilde{R}_z\rangle_s\chi_8(\nu). \qquad (18)$$

Here

$\chi_1(\nu)=\frac{\bar{\Gamma}_\parallel}{\bar{\Gamma}_\parallel^2+\nu^2}$, $\chi_2(\nu)=\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-\omega)^2}+\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu+\omega)^2}$,

$\chi_3(\nu)=\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu+2\bar{G}_R)^2}+\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-2\bar{G}_R)^2}$,

$\chi_4(\nu)=\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu+\omega-2\bar{G}_R)^2}+\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-\omega+2\bar{G}_R)^2}$,

$\chi_5(\nu)=\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu+\omega+2\bar{G}_R)^2}+\frac{\bar{\Gamma}_s}{\bar{\Gamma}_s^2+(\nu-\omega-2\bar{G}_R)^2}$,

$\chi_6(\nu)=\frac{(\nu+2\bar{G}_R)}{\bar{\Gamma}_s^2+(\nu+2\bar{G}_R)^2}-\frac{(\nu-2\bar{G}_R)}{\bar{\Gamma}_s^2+(\nu-2\bar{G}_R)^2}$,

$\chi_7(\nu)=\frac{(\nu+\omega-2\bar{G}_R)}{\bar{\Gamma}_s^2+(\nu+\omega-2\bar{G}_R)^2}-\frac{(\nu-\omega+2\bar{G}_R)}{\bar{\Gamma}_s^2+(\nu-\omega+2\bar{G}_R)^2}$,

$\chi_8(\nu)=\frac{(\nu+\omega+2\bar{G}_R)}{\bar{\Gamma}_s^2+(\nu+\omega+2\bar{G}_R)^2}-\frac{(\nu-\omega-2\bar{G}_R)}{\bar{\Gamma}_s^2+(\nu-\omega-2\bar{G}_R)^2}$,

$\bar{\Gamma}_s=4\bar{\Gamma}_0+\bar{\Gamma}_++\bar{\Gamma}_-$, $\Gamma_\parallel=2(\bar{\Gamma}_-+\bar{\Gamma}_+)$, $\langle\tilde{R}_z\rangle_s=\frac{\bar{\Gamma}_--\bar{\Gamma}_+}{\bar{\Gamma}_-+\bar{\Gamma}_+}$

In **Fig.1** we plot the squeezing spectrum for certain parameters of interest. Particularly, squeezing occurs for negative values and broader squeezing ranges are taking place because of permanent dipoles (compare the dashed and solid lines in **Fig. 1**, see also [6]).

IV. Conclusion.

We have discussed squeezing effects in resoance fluorescence processes of laser pumped two-level systems possessing permanent dipoles. We have found addictional quantum fluctuations domains which are due to permanent dipoles.

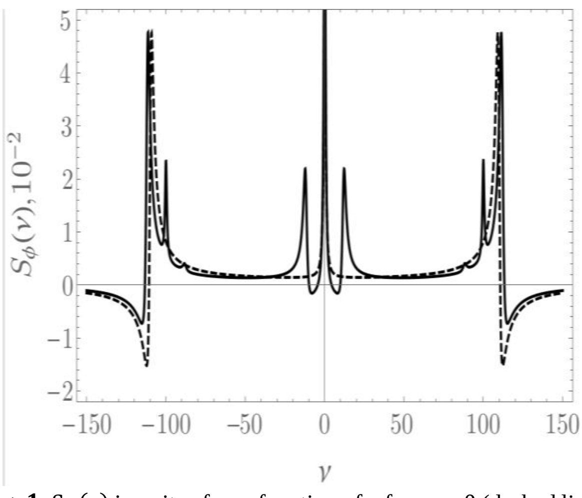

Fig. 1. $S_\varphi(\nu)$ in units of γ as function of ν for $g=0$ (dashed line) and $g=16$ (solid line). Other parameters are $\varphi=-\frac{\pi}{4}$, ω = 100, $q=\frac{\Delta}{2\Omega}$ = 0.7, Ω = 45.

Bibliography

[1] D. F. Walls, G. J. Milburn, "Quantum Optics", Springer, 2008.

[2] C. Gerry, P. Knight, "Introductory Quantum Optics ", Cambridge Univeristy Press, 2005.

[3] D. F. Walls, P. Zoller, "Reduced Quantum Fluctuations in Resoance Fluorescne", Physical Review Letters, Volume 47, Number 10, pp. 709-711, 1981.

[4] M. Macovei, M. Mishra, C. H. Keitel, "Population inversion in two-levle systems possessing permanent dipoles", Physical Review A, 92, pp. 013846-1 – 013846-5, 2015.

[5] F. Oster, C. H. Keitel, M. Macovei, "Generation of correlated photon pairs in different frequency ranges", Physical Review A, 85, pp.063814-1 – 063814-5, 2012.

[6] M. A. Anton, S. Maede-Razavi, F.Carreno, I.Thanopulos, and E.Paspalakis, "Optical and microwave control of resonace fluorescence and squeezing spectra in a polar molecule", Physical Review A 96, pp. 063812-1 – 063812-15, 2017.

[7] O. V. Kibis, G.Y.Slepyan, S.A Maksimenko, and A. Hoffmann, "Matter Coupling to Strong Electromagnetic Fields in Two-Level Quantum Systems with Broken Inversion Symmetry", Physical Review Letters, 102, 023601, 2009.

[8] G.Y.Kryuchkyan, V. Shahnazaryan, O. V. Kibis, and I. A. Shelykh, "Resonance fluorescence from an asymmetric quantum dot dressed by a bichromatic electromagnetic field", Physical Review A 95, 013834, 2017.

[9] A. Mirzac, M. Macovei, "Squeezing in Fluorescence Spectrum of Pumped Molecular Systems with Permanent Dipole", Humboldt Kolleg: Multidisciplinarity in Modern Science for the Benefit of Society (open workshop). Program & Abstracts, pp. 58, September 2017.

[10] A. Mirzac, "Resonance Fluorescence Spectrum in Pumped Molecular Systems with Permanent Dipole", Tendinte Contemporane ale Dezvoltarii Stiintei: Viziuni ale Tinerilor Cercetatori, Volumul I, pp. 79 – 84, Conferinta Stiintifica a Doctoranzilor (cu participare internationala), editia a VI-a, Chisinau, 15 iunie 2017.

Q. V. Lawande, S. V. Lawande, "Squeezing in cooperative resonance fluorescence", Physical Review A 38, pp. 800 – 807, 1988.