Pis'ma v ZhETF, vol. 100, iss. 7, pp. 480 – 483

Resonance fluorescence of localized multiatomic ensembles

L. V. Il'ichov[+1)], *A. V. Taychenachev*[*×]

[+]*Institute of Automation and Electrometry SB RAS, 630090 Novosibirsk, Russia*

[*]*Institute of Laser Physics SB RAS, 630090 Novosibirsk, Russia*

[×]*Novosibirsk State University, 630090 Novosibirsk, Russia*

Submitted 4 August 2014

The issue of fluorescing multiatomic ensembles is addressed in a somewhat unconventional fashion. We are concerned with the case of an ensemble being localized within a volume small compared to the wavelength. Collective phenomena in resonance fluorescence make its analytical description problematic. We employ the representation due to Holstein and Primakoff of atomic pseudo-spin operators. When the dispersion of the number of excited atoms is small, the kinetic equation turns into that for an effective quantum photonic mode. The new equation is nonlinear. Its stationary solution can easily be found nevertheless. The motion of two fluorescing ensembles is considered as well. Their interaction appears to have geometric nature.

DOI: 10.7868/S0370274X14190023

Introduction. Novel techniques of laser ultracooling enable one to assemble and manipulate quantum multiatomic systems [1]. The variety of physical processes these systems are involved at, challenges seriously our ability to provide their relevant theoretical description. Interatomic forces pose indisputably the main problem. The adequate approach to cooperative phenomena in emitting atomic ensembles is hardly less problematic. When such an ensemble is localized so that the distance between any pair of atoms is less than the wavelength λ, no information is available about the atom responsible for any given act of spontaneous emission. Various possible emission scenarios interfere therefore, which results in a highly entangled state in the process of spontaneous emission. In the seminal work [2] Dicke studied peculiarities of collective spontaneous emission. It is worth to stress that no direct interaction between atoms is needed to create the entanglement. The last is generated by the irreversible process of spontaneous emission of all atoms into common environment.

Let us take a localized ensemble of N two-level atoms with excited state $|e\rangle$ and ground state $|g\rangle$. The collective spontaneous decay of initially prepared state $|e\rangle^{\otimes N}$ terminates naturally in $|g\rangle^{\otimes N}$. If the spontaneous decay is accompanied by excitation due to interaction of atoms with an external monochromatic field, one may expect to observe some non-trivial stationary kinetic state of the ensemble. There will be a fraction of excited atoms. This is the regime of resonance fluorescence.

The (pseudo)spin model of atomic ensemble is useful when treating the resonance fluorescence. In the case of one atom the operators $\hat{S}_0 = (|e\rangle\langle e| - |g\rangle\langle g|)/2$, $\hat{S}_+ = |e\rangle\langle g|$ and $\hat{S}_- = |g\rangle\langle e|$ have the following commutation relations:

$$[\hat{S}_0, \hat{S}_\pm] = \pm\hat{S}_\pm,\ [\hat{S}_+, \hat{S}_-] = 2\hat{S}_0. \tag{1}$$

The operators $\hat{S}_0$ and $\hat{S}_\pm$ appear to be the generator of $SU(2)$ group, so revealing the equivalence of two-level atom and spin-half system. In terms of angular momentum notations $|1/2, 1/2\rangle = |e\rangle$ and $|1/2, -1/2\rangle = |g\rangle$. The kinetic equation of resonantly fluorescing atom reads

$$\partial_t \hat{\varrho}_t = -i\Delta[\hat{S}_0, \hat{\varrho}_t] - i\Omega[\hat{S}_+ + \hat{S}_-, \hat{\varrho}_t] + \tag{2}$$

$$+ 2\Gamma\hat{S}_-\,\hat{\varrho}_t\hat{S}_+ - \Gamma\{\hat{S}_+\hat{S}_-, \hat{\varrho}_t\},$$

where $\Delta = \omega_0 - \omega$ is the frequency detuning (ω_0 is the frequency of atomic transition and ω is that of classical monochromatic field), Ω is the Rabi frequency, and Γ is the rate of spontaneous decay, $\{\ldots\}$ stands for anticommutator. This equation as well as the (pseudo)spin model is equally applicable to N-atomic localized system. Really, due to system's localization neither interaction with external light, nor spontaneous decay can change the symmetry of N-atomic state with respect to permutation of particles. In particular, the state being initially invariant under any permutation (e.g. $|e\rangle^{\otimes N}$) possesses this property at all times. In this case the

[1)]e-mail: leonid@iae.nsk.su

ensemble of atoms is equivalent to $N/2$-spin system: $|N/2, N/2\rangle = |e\rangle^{\otimes N}$, ..., $|N/2, -N/2\rangle = |g\rangle^{\otimes N}$. Here the second quantity in the kets is the projection of the "spin" onto the fixed energy-like direction in the effective 3D space. We must point that no dipole-dipole interaction is present in (2). One may formally justify this option taking the limit $d \to 0$, $\Omega = Ed = \text{const}$ (d is the atomic dipole moment, E is the amplitude of light field). We also neglect the motion of atoms. These simplifying assumptions are similar to those made in Tavis–Cummings model [3, 4].

The expression for the stationary kinetic state of one fluorescing atom can easily be established. Not so for multiatomic ensemble. The stationary solution $\hat{\varrho}^{(st)}$ to (2) is an element of the universal enveloping algebra [5] generated by $\hat{S}_0$ and $\hat{S}_\pm$. In the case of $N = 1$ this algebra is merely $span\{\hat{1}, \hat{S}_0, \hat{S}_\pm\}$. The case $N = 2$ (unit (pseudo)spin) is more cumbersome but still tractable in contrast to the situation $N \gg 1$. In the last case the problem of $\hat{\varrho}^{(st)}$ evaluation seems to lie beyond a handmade analytical treatment. An alternative approach is needed. It must naturally be approximate.

Model. It would be instructive to make some assertions about the nature of stationary solution to (2). Note that the loss $\sim \Gamma \hat{S}_- \hat{\varrho} \hat{S}_+$ of atomic system's energy competes against the income $\sim \Omega(\hat{S}_+ \hat{\varrho} - \hat{\varrho}\hat{S}_-)$. Let us estimate the competing magnitudes for $\hat{\varrho} = |S, M\rangle\langle S, M|$. For $M = \pm(S-1)$ and $S \gg 1$ we get respectively $\sim \Gamma S$ and $\sim \Omega\sqrt{S}$ whereas for $M \sim 0$ these magnitudes are $\sim \Gamma S^2$ and $\sim \Omega S$. Because of rapid collective decay in the last case, steady kinetic states are more likely to survive near the poles of Bloch sphere (for $|M| \sim S$) rather than near its equator. This conclusion will be useful later on.

The so-called Holstein–Primakoff representation [6] is known for the operators (1) which act in the space of states with definite spin value $span\{|S, M\rangle : M = -S, -S+1, \ldots, S\}$:

$$\hat{S}_0 = \hat{a}^\dagger \hat{a} - S,$$

$$\hat{S}_+ = \hat{a}^\dagger \sqrt{2S - \hat{a}^\dagger \hat{a}}, \ \hat{S}_- = \sqrt{2S - \hat{a}^\dagger \hat{a}}\, \hat{a}, \qquad (3)$$

where $\hat{a}$ and $\hat{a}^\dagger$ obey the commutation relation $[\hat{a}, \hat{a}^\dagger] = 1$ of Bose-type. Let us assume that under some conditions the dispersion of $\hat{n} \equiv \hat{a}^\dagger \hat{a}$ in the state $\hat{\varrho}_t$ is small, so that one may approximate $\hat{S}_\pm$:

$$\hat{S}_+ \simeq \sqrt{2S - \langle n\rangle_t}\, \hat{a}^\dagger, \ \hat{S}_- \simeq \sqrt{2S - \langle n\rangle_t}\, \hat{a}. \qquad (4)$$

Here $\langle n\rangle_t = \operatorname{Tr} \hat{n}\hat{\varrho}_t$. The kinetic equation (2) takes the form (with $2S = N$)

$$\partial_t \hat{\varrho}_t = -i\Delta[\hat{a}^\dagger \hat{a}, \hat{\varrho}_t] - i\Omega\sqrt{N - \langle n\rangle_t}[\hat{a} + \hat{a}^\dagger, \hat{\varrho}_t] + \qquad (5)$$

$$+ 2\Gamma(N - \langle n\rangle_t)\hat{a}\, \hat{\varrho}_t \hat{a}^\dagger - \Gamma(N - \langle n\rangle_t)\{\hat{a}^\dagger \hat{a}, \hat{\varrho}_t\}.$$

This is the equation for an imaginary photonic mode interacting with a classical (quasi)harmonic dipole $\propto \Omega\sqrt{N - \langle n\rangle_t}$ and subjected to loss of quanta with the rate $\Gamma(N - \langle n\rangle_t)$.

Eq. (5) is non-linear. Nevertheless, its steady-state solution is straightforward. This is the Glauber coherent state

$$\hat{\varrho}^{(st)} = |\alpha\rangle\langle\alpha|, \qquad (6)$$

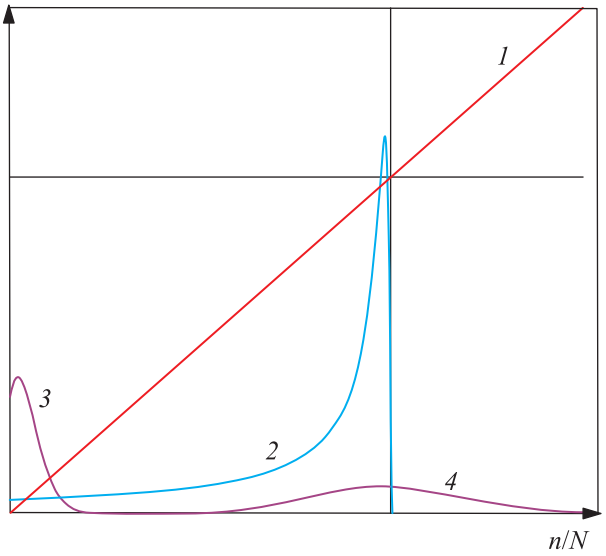
where

$$\alpha = -\frac{i\Omega\sqrt{N - \langle n\rangle^{(st)}}}{\Gamma(N - \langle n\rangle^{(st)}) + i\Delta}. \qquad (7)$$

The equation $\langle n\rangle^{(st)} = \operatorname{Tr} \hat{n}\hat{\varrho}^{(st)} = \langle\alpha|\hat{a}^\dagger \hat{a}|\alpha\rangle = |\alpha|^2$ delivers the following condition for $\langle n\rangle^{(st)}$:

$$\langle n\rangle^{(st)} = \frac{\Omega^2(N - \langle n\rangle^{(st)})}{\Gamma^2(N - \langle n\rangle^{(st)})^2 + \Delta^2}. \qquad (8)$$

This is a cubic equation in $\langle n\rangle^{(st)}$ (it turns into quadratic equation for $\Delta = 0$). Possible values of $\langle n\rangle^{(st)}$ are given by the intersections of LHS of (8), the line function of $\langle n\rangle^{(st)}$, and its RHS, the dispersion-like function. Depending on the values Ω/Γ, Δ/Γ, and N, there may be three, two, one or no one intersection points. Let us consider the case of three intersections (Figure). Two



Parameters: $N = 25$, $\Delta^2 = 0.2\Gamma^2$, $\Omega^2 = 25\Gamma^2$. The straight line *1* is the left-hand side of (8). The curve *2* is its right-hand side. The curve *3* is the distribution of excited atoms for the left, physically meaningful, intersection of *1* and *2*. The curve *4* is the same distribution for the right intersection

solutions of (8) (closest to 0 and to N) can be related to aforementioned expected steady-state solutions

of the kinetic equation. The first of these two points is evidently stable, for any shift towards greater values of $\langle n\rangle$ makes the collective decay stronger, which returns the point back. The same reasoning prompts us to conclude the unstable nature of the third intermediate point. Hence the greatest solution of (8) must be stable. The fact is yet insufficient for this solution being legitimate. The point is that the Poisson distribution of n in the state (6) for the greatest value of $\langle n\rangle^{(st)}$ is rather wide (see Figure). The unstable solution appears to be within the width. Hence the stable character of the greatest intersection point is in fact spurious. In addition the great width in n is inconsistent with the relevance of the assumption (4). A different situation obtains with the least solution to (8). The width of the Poisson distribution is much less in this case (the curve *3* at Figure). This particular value of $\langle n\rangle^{(st)}$ along with the corresponding $\hat{\varrho}^{(st)} = |\alpha\rangle\langle\alpha|$ have real chances to be close to their exact counterparts delivered by (2).

Discussion. There is an important case in which the solution for $\langle n\rangle^{(st)}$ is quite simple and straightforward. Really, if one may expect $\langle n\rangle^{(st)} \ll N$, (8) yields

$$\langle n\rangle^{(st)} \simeq \frac{N\Omega^2}{\Delta^2+\Omega^2+\Gamma^2N^2}. \tag{9}$$

If RHS of this expression is great (this may be in the case $N \gg 1$), the dispersion of n in the corresponding coherent state is of the order of $\sqrt{\langle n\rangle^{(st)}}$ [4]. This quantity is by all means negligible when compared with $N - \langle n\rangle_t$ in (5) for $\langle n\rangle_t$ closed to $\langle n\rangle^{(st)}$.

The proposed approach can shed some light on the peculiarities of relative motion of two localized coherent fluorescing ensembles of atoms, 1 and 2. In what follows we will address them as "drops". Assume the translational motion of the drops to be much slower then their internal radiation-induced evolution. Hence for this evolution the positions $\mathbf{r}_1$ and $\mathbf{r}_2$ of the drops may be treated as parameters. We consider the plain pumping wave which inserts the proper $\mathbf{r}$-dependence to the Rabi frequencies. The master equation for the state operator of the drops reads:

$$\begin{aligned}\partial_t\hat{\varrho}_t = &-i\Delta\sum_{i=1,2}[\hat{a}_i^\dagger\hat{a}_i,\hat{\varrho}_t] - \\ &- i\Omega\sum_{i=1,2}\sqrt{N_i-\langle n_i\rangle_t}[\hat{a}_i^\dagger e^{-i\mathbf{k}\mathbf{r}_i}+\hat{a}_i e^{i\mathbf{k}\mathbf{r}_i},\hat{\varrho}_t] + \\ &+\Gamma\sum_{i=1,2}(N_i-\langle n_i\rangle_t)\left(2\hat{a}_i\,\hat{\varrho}_t\,\hat{a}_i^\dagger-\{\hat{a}_i^\dagger\hat{a}_i,\hat{\varrho}_t\}\right)+ \\ &+\gamma(r)\sum_{i\neq j}\sqrt{(N_i-\langle n_i\rangle_t)(N_j-\langle n_j\rangle_t)}\times \\ &\times\left(2\hat{a}_i\,\hat{\varrho}_t\,\hat{a}_j^\dagger-\{\hat{a}_j^\dagger\hat{a}_i,\hat{\varrho}_t\}\right).\end{aligned} \tag{10}$$

The third line stands for spontaneous emissions with unambiguous identification of the drop responsible for the emitted photon. On the contrary, the last line takes account of the interference of the both scenarios; the factor $\gamma(r) = \Gamma\sin(kr)/kr$ appears due to integration over all possible directions of spontaneous emissions; this rate depends on the length of $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$. We introduce also the center of mass of the two drops, so that

$$\mathbf{r}_1 = \mathbf{R} + \frac{m_2}{M}\mathbf{r},\ \ \mathbf{r}_2 = \mathbf{R} - \frac{m_1}{M}\mathbf{r}.$$

Here $M = m_1 + m_2$. The steady-state solution to (10) is the product of coherent states:

$$|e^{-i\mathbf{k}\mathbf{R}}\alpha_1(\mathbf{r})\rangle_1\langle e^{-i\mathbf{k}\mathbf{R}}\alpha_1(\mathbf{r})|\otimes|e^{-i\mathbf{k}\mathbf{R}}\alpha_2(\mathbf{r})\rangle_2\langle e^{-i\mathbf{k}\mathbf{R}}\alpha_2(\mathbf{r})|, \tag{11}$$

where

$$\begin{aligned}\alpha_1(\mathbf{r}) = &\\ = -i\Omega\sqrt{N_1-\langle n_1\rangle}&\Big\{[\Gamma(N_2-\langle n_2\rangle)+i\Delta]e^{-i\mathbf{k}\mathbf{r}m_2/M}- \\ -\gamma(r)(N_2-\langle n_2\rangle)e^{i\mathbf{k}\mathbf{r}m_1/M}&\Big\}\Big\{[\Gamma(N_1-\langle n_1\rangle)+i\Delta]\times \\ \times[\Gamma(N_2-\langle n_2\rangle)+i\Delta]-\gamma(r)^2(N_1-\langle n_1\rangle)&(N_2-\langle n_2\rangle)\Big\}^{-1}.\end{aligned} \tag{12}$$

The expression for $\alpha_2(\mathbf{r})$ is similar and can be get from (12) by interchange of 1 and 2 indices. The mean steady-state values of excited atoms in the drops appear as the solutions to $\langle n_1\rangle = |\alpha_1(\mathbf{r})|^2$ and $\langle n_2\rangle = |\alpha_2(\mathbf{r})|^2$. The important point for what follows is that the internal state of the drops (11) is pure. In the case of pure *total* state it may hence be written as

$$\begin{aligned}|\Psi\rangle = \int\psi(\mathbf{r}_1,\mathbf{r}_2)|\mathbf{r}_1,\mathbf{r}_2\rangle\otimes \\ \otimes|e^{-i\mathbf{k}\mathbf{R}}\alpha_1(\mathbf{r})\rangle_1\otimes|e^{-i\mathbf{k}\mathbf{R}}\alpha_2(\mathbf{r})\rangle_2 d^3\mathbf{r}_1 d^3\mathbf{r}_2.\end{aligned} \tag{13}$$

It is worth to note that both the external and internal states of the pair of drops are generally mixed due to entanglement between the translational and internal degrees of freedom, and the function $\psi(\mathbf{r}_1,\mathbf{r}_2)$ should not be confused with a wave function. Rapid radiational processes have already been taken into account in the state (13). Its slow evolution is governed by kinetic Hamiltonians of the drops[2)]:

$$i\partial_t|\Psi\rangle = \left(\frac{\hat{p}_1^2}{2m_1}+\frac{\hat{p}_2^2}{2m_2}\right)|\Psi\rangle. \tag{14}$$

[2)]For the sake of simplicity we take into account no external potentials. In real situation these appear as an optical lattice, for example, created by some non-resonant field.

The resulting equation for $\psi(\mathbf{r}_1, \mathbf{r}_2)$ written as a function of $\mathbf{R}$ and $\mathbf{r}$ reads

$$i\partial_t\psi(\mathbf{R},\mathbf{r}) = -\frac{1}{2M}\Big[(\partial_{\mathbf{R}} - i\mathbf{k})^2 - 2k^2\Big]\psi(\mathbf{R},\mathbf{r}) - \\ - \frac{1}{2m}\Big\{[\partial_{\mathbf{r}} - i\mathbf{A}(\mathbf{r})]^2 + V(\mathbf{r})\Big\}\psi(\mathbf{R},\mathbf{r}). \qquad (15)$$

Here the vector and scalar potentials in drops' relative motion appear:

$$\mathbf{A}(\mathbf{r}) = \mathrm{Im}\sum_{i=1,2}\alpha_i^*(\mathbf{r})\partial_{\mathbf{r}}\alpha_i(\mathbf{r}), \qquad (16)$$

$$V(\mathbf{r}) = -\sum_{i=1,2}|\partial_{\mathbf{r}}\alpha_i(\mathbf{r})|^2 - \mathbf{A}^2(\mathbf{r}). \qquad (17)$$

The potentials have pure geometric origin. There is a resemblance with geometric potentials in the motion of atoms *as individual objects* under coherent population trapping in optical fields with non-trivial configurations [7]. In our case the geometric potentials govern the *relative* motion of the drops in the plain wave.

Resuming, there proposed the approximate approach to the description of resonance fluorescence of localized multiatomic ensembles. Steady-state of the fluorescing ensemble, Glauber coherent state, is quite simple in this approach. In a sense the greater number of atoms constitutes the ensemble, the more relevant the approach is. It may find applications in many fields. In particular, the approach makes it possible to extend the description of feedback phase-switching from the case of one and two fluorescing atoms [8] to multiatomic case.

One of the authors (I.L.) acknowledges the support from RFBR (grant #12-02-01130), the Presidium of SB RAS, and the RAS Physical Department's program "Fundamental optical spectroscopy and applications".

1. S. Haroche and J.-M. Raimonde, *Exploring the Quantum*, Oxford University Press (2006).
2. R. H. Dicke, Phys. Rev. **93**, 99 (1954).
3. M. Tavis and F. W. Cummings, Phys. Rev. A **170**, 379 (1968).
4. A. B. Klimov and S. M. Chumakov, *A Group-Theoretical Approach to Quantum Optics*, WILEY-VCH Verlag GmbH and Co (2009).
5. J. E. Humphreys, *Introduction to Lie Algebras and Representation Theory*, Springer (1978).
6. T. Holstein and H. Primakoff, Phys. Rev. A **58**, 1098 (1940).
7. A. V. Taychenachev, A. M. Tumaikin, and V. I. Yudin, Laser. Phys. **2**, 575 (1992); R. Dum and M. Olshanii, Phys. Rev. Lett. **76**, 1788 (1996); P. M. Visser and G. Nienhuis, Phys. Rev. A **57**, 4581 (1998).
8. V. A. Tomilin and L. V. Il'ichov, JETP Lett. **94**, 676 (2011); JETP **116**, 181 (2013); Doklady Physics **58**, 413 (2013).