An application of the spectral Radon–Nikodym approach to fluorescence dynamics study of cold atomic cloud ensemble

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Abstract. A fluorescence dynamics of a cold atomic cloud, laser–cooled to a very low temperature, optically excited by a short laser impulse is calculated by applying non–stationary Schrödinger equation to the system of atoms and electromagnetic field. The solution gives fluorescence time–relaxation dependence, with relaxation time distribution depending on the system itself and external electromagnetic field applied. The distribution of the relaxation rates changes, when an external electric field affecting the atomic ensemble is applied. To analyze the relaxation process a novel Radon–Nikodym approach is applied: the relaxation curve is not piece–wise interpolated, but instead is converted to a matrix, the eigenvalues of which provide relaxation rate distribution. In contrasts to the random matrix theory, where the weights, corresponding to an eigenvalue, are considered equal, in this work the weight, corresponding to an individual eigenvalue, is determined by the Lebesgue integral quadrature weights.

1. Introduction

One of the most expressive multi–particle quantum systems is a cloud of atoms (cesium, rubidium) trapped in a magneto-optical trap (MOT). Such atomic ensembles are considered to be very perspective systems in quantum informatic and frequency standarts [1–3]. The goal of this work is a theoretical analysis of fluorescence temporal dynamics of ensembles of cold atoms, excited by a laser pulse. This allows us to analyze the physical properties of atomic ensembles [4–10], and also enables the control and management of their optical properties by applying external electric and magnetic fields. The properties of atomic clouds in traps are calculated using the model of fixed scatterers and allow a consistent quantum microscopic description based on the solution of the nonstationary Schrödinger equation [11–20]. This equation is solved for a unified system consisting of atoms and an electromagnetic field. Such an approach makes it possible, within the framework of a single formalism, to take into account: the interaction of atoms with external electromagnetic radiation, resonant dipole-dipole interaction between atoms, and spontaneous decay of their excited states.

The analysis of the temporal dynamics of the fluorescence of an ensemble of cold atoms is an effective way of experimentally controlling such clouds. Specific computer calculations were performed for atomic clouds having a spherically symmetrical Gaussian distribution of atoms. The problem was averaged over an ensemble of random configurations of atoms. The radius of the cloud $R = 10 \lambda$ and the concentration of atoms $n_0$ satisfy the relation $n_0 \lambda^3 = 0.2$, where $\lambda = 1/k$ (the wave vector of the incident radiation). At such a concentration, the ensemble of cold atoms is a single large cluster in which collective effects are strong. The excitation pulse was considered circularly polarized. The duration of the exciting pulse $\tau_L$ was...
chosen to be much less than the lifetime $\tau_a$ of the excited states $\tau_L/\tau_a = 0.1$. This choice of the $\tau_L$ makes it possible to effectively excite such collective states in a fairly wide spectral range. The carrier frequency of the exciting pulse was set equal to the frequency of the atomic transition of an isolated atom. The temporal behavior of the fluorescence signals was calculated for different angles $\theta = \{\pi/20, \pi/4, \pi/2, \pi\}$, measured from the direction of the incident wave. For each angle, the fluorescence was analyzed both with conserved $H \parallel H$ and changing $H \perp H$ helicity. The paper will consider the temporal behavior of fluorescence, both in the absence of an external electric field $E = 0$ and when a strong external field applied $E_0 \neq 0$. A strong field means that the level splitting $\Delta$ caused by the electric field satisfies the relation $\Delta \gg \hbar/\tau_a$.

For all the cases considered, a complex time dependence of the fluorescence signals was obtained. The fluorescence intensity $I(t)/I_0$ (the fluorescence intensity at $t = 0$) decreases with time according to a multiexponential law. At short times $t/\tau_a \leq 1$, the decay rate exceeds the rate of spontaneous decay of free atoms. In this case, superradiance is observed. At large times $t/\tau_a \gg 1$, long–lived collective states make the main contribution, and a sub–radiation effect is observed. In figure 1, $z(t) = \ln (I(t)/I_0)$ is plotted on the logarithmic scale. Depending on the dimensionless time $t/\tau_a$, the dependence of the total intensity $I_\perp + I_\parallel$ at the angle $\theta = \pi/2$ is plotted. For such a case, the difference in the temporal behavior of the intensities in the case of an external electric field $E_0 = 0$ and $E_0 \neq 0$ is most pronounced.

Analyzing the figure 1, we can conclude that the most long–lived collective excitations in ensembles of cold atoms make the main contribution to the long–term fluorescence relaxation. The fluorescence decay dynamics at large times is associated with long–lived states, which are the collective behavior of a dense ensemble of cold atoms. The reason for their appearance is that when atoms approach each other, collective, quasimolecular states are formed due to the interatomic interaction. At such high densities $n_0 a^3 = 0.2$, the cluster of cold atoms behave like a single quasi-molecule. The inclusion of the electric fields makes the system anisotropic. The external field due to the Stark effect shifts some of these states. In addition, the external field modifies the collective Lamb shift caused by the interatomic interaction. All this, at large times $t/\tau_a \gg 1$, leads to the reduction of fluorescence intensity with the electric field $E_0 \neq 0$ compared to the one in the absence of the electric field $E_0 = 0$.

Figure 1. The $z(t) = \ln (I(t)/I_0)$ dependence on the dimensionless time $t/\tau_a$ with and without electric fields.
Figure 2. Eigenvalues distribution of $f = z$ for $E = 0$ and $E \neq 0$ cases with the Lebesgue quadrature [25] weights $w[i] = \langle \psi[i] \rangle^2$, $n = 50$, and the measure $d\mu = dt$. In this case weights sum is equal to total interval length. The minimal/maximal eigenvalue can be used [21] as an estimator of the global min/max of an observable $f$ in (5) problem.

For relaxation process analysis the spectral Radon–Nikodym approach [21–23] has been used. The main idea is to consider not a traditional interpolation of an observable $f$ as a linear superposition of basis functions:

$$ f \approx \sum_{k=0}^{n-1} \beta_k Q_k(x), \quad (1) $$

but instead to introduce a wavefunction $\psi(x)$ as a linear superposition of basis functions, then to average an observable $f(x)$ with the $\psi^2(x)d\mu$ weight:

$$ \psi(x) = \sum_{j=0}^{n-1} \alpha_j Q_j(x), \quad (2) $$

$$ f_\psi = \frac{\langle \psi \mid f \mid \psi \rangle}{\langle \psi \mid \psi \rangle} = \sum_{j=0}^{n-1} \alpha_j \langle Q_j \mid f \mid Q_j \rangle \alpha_j, \quad (3) $$

With a positively defined matrix $\langle Q_j \mid Q_k \rangle$ generalized eigenvalue problem:

$$ \begin{align*}
  \langle f \mid \psi[i] \rangle &= \lambda[i] \langle \psi[i] \rangle \\
  \sum_{k=0}^{n-1} \langle Q_j \mid f \mid Q_k \rangle \alpha_k[i] &= \lambda[i] \sum_{k=0}^{n-1} \langle Q_j \mid Q_k \rangle \alpha_k[i], \quad (5) \\
  \psi[i](x) &= \sum_{k=0}^{n-1} \alpha_k[i] Q_k(x) \quad (6)
\end{align*} $$

has a unique solution. Found eigenfunctions to be normalized as $\langle \psi[i] \mid \psi[j] \rangle = \delta_{ij}$. Then $\langle \psi[i] \mid f \mid \psi[j] \rangle = \lambda[i] \delta_{ij}$; $\sum_{l,m=0}^{n-1} \alpha_l[i] \langle Q_l \mid Q_m \rangle \alpha_m[j] = \delta_{ij}$; and $\lambda[i] = \left\langle \left(\psi[i] \right)^2 \right\rangle \left(\left\langle \psi[i] \right\rangle \right)^2$. All the temporal dynamics
information contains in two matrices $\langle Q_i | f | Q_k \rangle$ and $\langle Q_j | Q_k \rangle$, not in vectors $\langle f | Q_k \rangle$ like in Fourier, least squares, and the other norm–minimizing approaches. Note that as an $f$ we can take either $z(t)$ or $dz/dt$, in both situation the matrix $\langle Q_j | f | Q_k \rangle$ can be calculated directly from sample; in all the calculations below $f = z(t)$ is taken as an observable. A numerical solution of generalized eigenvalue problem can be performed using subroutines from several software packages. Obtained eigenvalues $\lambda^{(l)}$ model the distribution of $f$, a histogram can be plotted. But what the weight $w^{(l)}$ to correspond to an individual eigenvalue? There are two possible options:

- Equal weights $w^{(l)} = 1$ : $n = \sum_{i=0}^{n-1} w^{(l)}$. This approach is typically used in the random matrix theory [24]. One of the limitation of the equal weight approach is that the importance of different eigenvalues is typically not the same in practice.
- Lebesgue quadrature [25] weights $w^{(l)} = \langle \phi^{(l)} \rangle^2 ; \langle 1 \rangle = \sum_{i=0}^{n-1} w^{(l)}$. In this case the weights sum is equal the total measure, e.g. if one chose $dq = dt$ then the sum is equal to the interval total length. There is even more general approach, the density–matrix correlation [26], where for two problems [5], the cross–weights are calculated. For a single observable $f$ the Lebesgue quadrature weights $\langle \phi^{(l)} \rangle^2$ are invariant properties of the distribution of $f$.

Note that the relaxation curves (figure 1) are very similar in shape and differ only in the rate of the decay. The histograms in figure 2 of eigenvalues distribution for the case $E_0 = 0$ and $E_0 \neq 0$, are also similar in shape, but differ in the range of scatter of the eigenvalues. The generalized eigenvalue problem [5] is related to global optimization problem [21], and the minimal/maximal eigenvalue is a good estimator of global min/max. Thus, the range of eigenvalues for each constructed histogram is a criterion for the difference between the two curves shown in figure 1. If the relaxation processes had different scenarios of intensity behavior with time, this circumstance would lead to a different type of eigenvalues distribution.

2. Conclusion

In the study of collective effects of photons exchange between atoms in dense ensembles of cold atoms, the study of time dependence of the fluorescence intensity is of great importance. The fluorescence dynamics of a dense ensemble of cold atoms was calculated both in the case of the absence of an external electric field and in the case of a strong electric field. The analysis of the behavior of fluorescence in time is performed by applying the Radon-Nikodym method to the analysis of the eigenvalues of the signal matrix. The approach allows us to determine the presence of clusters formed in dense ensembles of cold atoms [27–33].

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