Influence of dipole-dipole interaction on collective spontaneous emission phenomena

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Abstract

In this work we study the effects of dipole-dipole interactions in systems, which possibly may exhibit superradiant behaviour. Starting with the Born-Markov approximation, we obtain equations for mean values of atomic operators for different regimes of superradiance. We simulate these equations numerically and obtain solutions for atomic observables.

Contents

1 Introduction

Collective coherent spontaneous emission, or superradiance, has been a topic of interest for many researchers from various fields of quantum optics related to light-matter interactions. Due to unique features and possibilities provided by such a class of phenomena, it has been thoroughly studied for a variety of systems using different approaches, from the classical Maxwell equations and many-body physics methods to purely quantum descriptions.

However, the influence of elastic dipole-dipole interactions of atoms, which usually are not taken into account in the description of collective emission, still remains an unsolved issue. At first sight, due to dependence of the dipole-dipole interaction on interatomic distances, each pair of atoms obtains a frequency shift which leads to dephasing and subsequent ruining of superradiance process. Thus samples with large dipole-dipole interactions (DDI) such as dense clouds of Rydberg atoms should not superradiate. Indeed, some groups have reported the absence of superradiance. However, other groups managed to observe superradiance directly in such systems [1]. Therefore it is required to work out a way for correct inclusion of the dipole-dipole interaction and study its influence on the collective spontaneous emission phenomena.

In this work we use the approach based on the Born-Markov Master equation [2]. The main idea is that we treat dipole-dipole interaction as a result of interaction between every individual atom and quantized electromagnetic field. In our numerical simulations we rely on the experimental parameters from [1]. We show that the Born-Markov approach is applicable for such experimental conditions, and then obtain equations for the mean values of atomic operators. Since it is not possible to obtain a rigorous closed set of equations, we assume factorization of operators of different atoms, in other words we work in a semi-classical regime. Relying on the experimental parameters, we simulate obtained equations numerically.

2 Theoretical approach

Starting point of our research is the Master equation for a reduced density matrix in the Born-Markov approximation. Consider a bipartite system, which consists of two interacting sub-systems, namely S and R, and is described by Hamiltonian $H = H_S + H_R + V$. In our case, such system is an atomic system coupled to a reservoir of quantized electromagnetic field modes. Typically we are only interested in the evolution of the atomic system, thus we need to conduct an exclusion of electromagnetic field. By tracing the density matrix of the whole system $\rho(t) = \rho_S(t) \otimes \rho_R(t)$, which satisfies von Neumann equation, over reservoir, one obtains a reduced density matrix of the atomic sub-system, which may be shown to satisfy the following equation in the Born-Markov approximation:

$$\dot{\rho}_S(t) = -\frac{1}{\hbar^2}\mathrm{Tr}_R \int_0^\infty d\tau \, [V(t), [V(t-\tau), \rho_S(t) \otimes \rho_R(0)]] \,. \tag{1}$$

Take a note that this equation is written in interaction picture. Born approximation typically implies neglecting time evolution of reservoir $\rho(t) \approx \rho_S(t) \otimes \rho_R(0)$, while Markov approximation neglects "memory" effects of evolution. Born approximation is valid only if the Markov approximation is, and applying both of them physically mean neglecting retardation effects [3].

2.1 Experimental parameters

In this section we will show that the Born-Markov approximation is applicable for describing the experimental sample from [1]. In this experiment, the sample has a pencil-shaped geometry with characteristic length $L = 15$ cm and radius 0.75 cm. This volume is filled by 10^7-10^9 Rydberg atoms of Barium, initially prepared in highly excited state $6s30p\,^1P_1$. Such densities and large values of dipole transition moments result in a significant dipole-dipole interaction between atoms.

Immediately following the excitation to the Rydberg state, a 10-ns mm-wave pulse triggers the superradiance. This pulse is on resonance with the $6s30p\,^1P_1 \rightarrow 6s28d\,^1D_2$ transition at 279.775 GHz, which corresponds to ≈ 1 mm wavelength. Therefore $L/\lambda \approx 150$, thus our system is not point-like.The triggering field has the energy ~ 0.3 pJ, which is about ten orders higher than the energy of single photon, therefore this pulse may be considered as classical field. Time retardation effects are of order $L/c = 5 \cdot 10^{-10}$ s. Another characteristic time is a time of spontaneous decay, which is given by:

$$\tau = \frac{3\hbar c^3}{4\omega^3 |\mu|^2}, \tag{2}$$

which for our system of Rydberg atoms ($|\mu| \sim 500$ D) is of order of 10 ms.

Duration of the pulse which triggers the superradiance is 10 ns. Such duration allows one to neglect both retardation effects and contribution of spontaneous decay into the process. Initial tipping angle of the Bloch-vector is $\pi/40$, and semi-classical approach will be a good approximation for such system.

2.2 Born-Markov Master equation

To specify the atomic system, we consider it as a set of motionless two-level systems distributed in space. In the framework of pseudospin formalism, we may write a full Hamiltonian with interaction taken in minimal coupling form as follows:

$$\begin{aligned} H = &\frac{\hbar\omega_0}{2}\sum_{i=1}^{N}\sigma_z^{(i)} + \hbar\sum_{\mathbf{k},\lambda}\omega_k a^\dagger_{\mathbf{k},\lambda}a_{\mathbf{k},\lambda} \\ &+i\hbar : \sum_{i=1}^{N}\sum_{\mathbf{k},\lambda}\left\{b_{\mathbf{k},\lambda}e^{i\mathbf{k}\mathbf{r}_i}a_{\mathbf{k},\lambda} + b^*_{\mathbf{k},\lambda}e^{-i\mathbf{k}\mathbf{r}_i}a^\dagger_{\mathbf{k},\lambda}\right\}\left\{\sigma_+^{(i)} - \sigma_-^{(i)}\right\} : . \end{aligned} \tag{3}$$

Note that this Hamiltonian contains only electrostatic interaction within individual atoms, i.e. no interaction between atoms is included. Also note that $\sim A^2$ shift is neglected since it is irrelevant for our task. Atomic transition frequency is ω_0, atom-field coupling $b_{\mathbf{k},\lambda} = \sqrt{\dfrac{2\pi}{V}}\dfrac{|e|\omega_0}{\sqrt{\hbar\omega_k}}(\boldsymbol{\mu}, \mathbf{e}(\mathbf{k},\lambda))$, where $\mathbf{e}(\mathbf{k},\lambda)$ denotes real field polarization vectors and $\mu = \langle e|\mathbf{r}|g\rangle$ is a real atomic transition polarization vector. Therefore coefficients $b_{\mathbf{k},\lambda}$ are real in such representation. In interaction picture corresponding to Hamiltonian (3):

$$V(t) = i\hbar : \sum_{\mathbf{k},\lambda,i} b_{\mathbf{k},\lambda}\left\{e^{i(\mathbf{k}\mathbf{r}_i - \omega_k t)}a_{\mathbf{k},\lambda} + e^{-i(\mathbf{k}\mathbf{r}_i - \omega_k t)}a^\dagger_{\mathbf{k},\lambda}\right\}\left\{\sigma_+^{(i)}e^{i\omega_0 t} - \sigma_-^{(i)}e^{-i\omega_0 t}\right\} :, \tag{4}$$

where colon operation denotes normal ordering of operators. Hence the Born-Markov Master equation for such system has a form:

$$\begin{aligned} \dot\rho_S(t) = -\sum_{\mathbf{k},\lambda,i,j}|b_{\mathbf{k},\lambda}|^2 e^{i\mathbf{k}(\mathbf{r}_i - \mathbf{r}_j)}\Bigg\{&\left(\frac{\pi}{c}\delta(k-k_0) - \frac{i}{c}\text{v.p.}\frac{1}{k-k_0}\right)\left[\sigma_+^{(i)}, \sigma_-^{(j)}\rho_S(t)\right] + \\ &+\left(\frac{\pi}{c}\delta(k+k_0) - \frac{i}{c}\text{v.p.}\frac{1}{k+k_0}\right)\left[\sigma_-^{(i)}, \sigma_+^{(j)}\rho_S(t)\right]\Bigg\} + h.c, \end{aligned} \tag{5}$$

where we utilized the Sokhotski–Plemelj theorem:

$$\int_0^\infty d\tau\, e^{-i(k\pm k_0)c\tau} = \frac{\pi}{c}\delta(k_0 \pm k) \mp \frac{i}{c}\text{v.p.}\frac{1}{k_0 \pm k}. \tag{6}$$

Omitting a principal value integral results in the Dicke superradiance regime [2], but we will keep this term as it accounts for non-resonant exchange of photons between atoms. Summation over polarization yields[1]

$$\sum_\lambda (\boldsymbol{\mu}, \mathbf{e}(\mathbf{k},\lambda)) = \mu^2 - \frac{(\boldsymbol{\mu}, \mathbf{k})^2}{k^2}. \tag{7}$$

[1]Here we used the identity which follows from complicity of polarization vectors:

$$\sum_\lambda e_\alpha(\mathbf{k},\lambda)\, e_\beta(\mathbf{k},\lambda) = \delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{k^2}.$$

The next step is replacing summation over wave-vectors by integration

$$\sum_{\mathbf{k}} = \frac{V}{(2\pi)^3} \int_0^\infty dk\, k^2 \int d\Omega(\mathbf{k}).$$

At this point it is possible to introduce the most general form of the dipole-dipole interaction (DDI) couplings, namely the inelastic part, responsible for collective decay processes:

$$F_{ij}(kr_{ij}) = \frac{3}{8\pi} \int d\Omega(\mathbf{k}) \left(1 - \frac{(\boldsymbol{\mu}, \mathbf{k})^2}{\mu^2 k^2}\right) e^{i\mathbf{k}(\mathbf{r}_i - \mathbf{r}_k)}, \tag{8}$$

(note that normalization is chosen so that $F_{ii}(0) = 1$) and the elastic part, which gives rise to collective lineshifts:

$$\Omega_{ij} = \frac{1}{\pi k_0} \text{v.p.} \int_{-\infty}^{+\infty} dk\, k \frac{F_{ij}(kr_{ij})}{k - k_0}. \tag{9}$$

Using the notation $F_{ij} \equiv F_{ij}(k_0 r_{ij})$, we rewrite the Born-Markov Master equation (5) as:

$$\begin{aligned} \dot{\rho}_S(t) = & -\gamma \sum_{i=1}^{N} \left(\left[\sigma_+^{(i)} \sigma_-^{(i)}, \rho_S(t) \right]_+ - 2\sigma_-^{(i)} \rho_S(t)\, \sigma_+^{(i)} \right) \\ & -\gamma \sum_{i \neq j} \left\{ (F_{ij} - i\Omega_{ij}) \left[\sigma_+^{(i)}, \sigma_-^{(j)} \rho_S(t) \right] + (F_{ij} + i\Omega_{ij}) \left[\rho_S(t)\, \sigma_+^{(i)}, \sigma_-^{(j)} \right] \right\}, \end{aligned} \tag{10}$$

where we have omitted the Lamb shift of each atom assuming that frequency transition is already renormalized.

2.3 Equations for mean values

From the Master equation for density matrix it is possible to obtain equations for mean values of atomic operators. Atomic coherences satisfy the following set of differential equations:

$$\left\langle \dot{\sigma}_+^{(k)}(t) \right\rangle = -\gamma \left\langle \sigma_+^{(k)}(t) \right\rangle + \gamma \sum_{i \neq k} (F_{ik} + i\Omega_{ik}) \left\langle \sigma_+^{(i)}(t)\, \sigma_z^{(k)}(t) \right\rangle, \tag{11}$$

and as for atomic populations:

$$\begin{aligned} \left\langle \dot{\sigma}_z^{(k)}(t) \right\rangle = & -2\gamma \left(1 + \left\langle \sigma_z^{(k)}(t) \right\rangle\right) - 2\gamma \sum_{j \neq k} (F_{kj} - i\Omega_{kj}) \left\langle \sigma_+^{(k)}(t)\, \sigma_-^{(j)}(t) \right\rangle \\ & -2\gamma \sum_{i \neq k} (F_{ik} + i\Omega_{ik}) \left\langle \sigma_+^{(i)}(t)\, \sigma_-^{(k)}(t) \right\rangle, \end{aligned} \tag{12}$$

note that this set of equations is not closed, so we assume that compositions of two operators of different atoms factorize:

$$\left\langle \sigma_{\pm,z}^{(k_1)}(t)\, \sigma_{\pm,z}^{(k_2)}(t) \right\rangle \approx \left\langle \sigma_{\pm,z}^{(k_1)}(t) \right\rangle \left\langle \sigma_{\pm,z}^{(k_2)}(t) \right\rangle, \quad k_1 \neq k_2, \tag{13}$$

which basically implies that we work in semi-classical regime.

3 Results

In this section we give the analytical and numerical results for different regimes of superradiance in the Born-Markov approximation.

3.1 Full angle integration

Coefficients (8), which describe inelastic part of DDI, have the following form in the case of full angle integration:

$$F_{ij} = \frac{3}{2}\sin^2\eta_{ij}\frac{\sin k_0 r_{ij}}{k_0 r_{ij}} + \frac{3}{2}(1-3\cos^2\eta_{ij})\cdot\left(\frac{\cos k_0 r_{ij}}{(k_0 r_{ij})^2} - \frac{\sin k_0 r_{ij}}{(k_0 r_{ij})^3}\right), \tag{14}$$

and hence coefficients (9), which account for imaginary part of the equations for atomic observables, are the following:

$$\Omega_{ij} = \frac{3}{2}\sin^2\eta_{ij}\frac{\cos k_0 r_{ij}}{k_0 r_{ij}} - \frac{3}{2}(1-3\cos^2\eta_{ij})\cdot\left(\frac{\sin k_0 r_{ij}}{(k_0 r_{ij})^2} + \frac{\cos k_0 r_{ij}-1}{(k_0 r_{ij})^3}\right). \tag{15}$$

Note that the full couplings of dipole-dipole interaction are complex and may be written as:

$$F_{ij} + i\Omega_{ij} = \nu_{ij}e^{-ik_0 r_{ij}} + \zeta_{ij}, \tag{16}$$

with complex coefficients:

$$\nu_{ij} = \frac{3}{2}\sin^2\eta_{ij}\frac{1}{(-ik_0 r_{ij})} + \frac{3}{2}(1-3\cos^2\eta_{ij})\cdot\left(\frac{1}{(k_0 r_{ij})^2} + \frac{1}{(-ik_0 r_{ij})^3}\right), \tag{17}$$

while ζ_{ij} resembles electrostatic dipole-dipole interaction between atoms:

$$\zeta_{ij} = \frac{3}{2}(1-3\cos^2\eta_{ij})\cdot\frac{i}{(k_0 r_{ij})^3}, \quad \zeta_{ij}^* = -\zeta_{ij}. \tag{18}$$

3.2 Integration in forth propagation direction

Assume that our atomic system is pencil-shaped and oriented along z-axis. It opens up a possibility to approximate integration over solid angle by a single term corresponding to small solid angle Δo in z direction, which results in the following form of the inelastic DDI coefficient (8):

$$F_{ik}(kr_{ik}) = \approx \frac{3\Delta o}{8\pi}\left(1-\frac{\mu_z^2}{\mu^2}\right)e^{ik(z_i-z_k)} \equiv \xi e^{ik(z_i-z_k)}, \tag{19}$$

and for elastic DDI (9):

$$\Omega_{ik} = i\xi\,\mathrm{sign}\,(z_i-z_k)\,e^{ik_0(z_i-z_k)}. \tag{20}$$

Consequently, the full DDI coupling shows the explicit effect of forth propagation:

$$F_{ij} + i\Omega_{ij} = \theta(z_i-z_j)\xi e^{ik_0(z_i-z_j)}, \tag{21}$$

where $\theta(z)$ is the Heaviside step function, which is 0 for $z<0$ and 1 for $z>1$. Here it is assumed that the atoms are ordered in such a way that $z_i > z_j$ if $i>j$.

3.3 Integration in forth and back propagation direction

Taking the backwards propagation into account in addition to forth direction propagation, the inelastic DDI coefficient takes a following form:

$$F_{ik}(kr_{ik}) \approx \frac{3\Delta o}{8\pi}\left(1-\frac{\mu_z^2}{\mu^2}\right)\left(e^{ik(z_i-z_k)}+e^{-ik(z_i-z_k)}\right) \equiv 2\xi\cos k|z_i-z_j|, \tag{22}$$

and for elastic coefficients:

$$\Omega_{ij} = -2\xi\sin k_0|z_i-z_j|, \tag{23}$$

thus, the full coupling is:

$$F_{ij}+i\Omega_{ij} = 2\xi e^{-ik_0|z_i-z_j|}. \tag{24}$$

By comparing (16), (21) and (24) it is possible to work out limit transition between these models. It allows to build-up a quasi-1D model from 3D model, which is important for angle Δo estimations.

3.4 Comparison of complex DDI and Dicke limit

We generated an atomic system of $N = 1000$ atoms randomly distributed in the cylinder with length $L = 2\lambda_0$ and radius $r = 0,001\lambda_0$. Polarization of atomic transition is orthogonal to the cylinder axis. The figure 1 shows the intensity calculated for different regimes of superradiance: complex DDI and Dicke limit. One can see that complex DDI regime exhibits intensity ringings, which were experimentally observed.

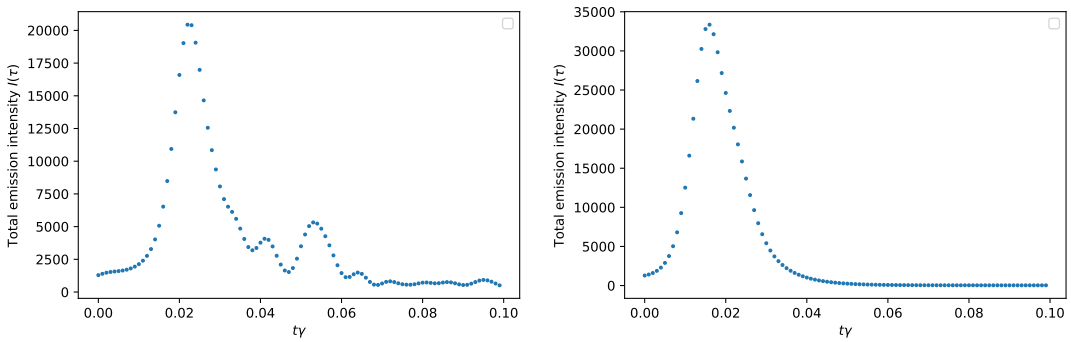

Figure 1: Intensity of emission in regime of complex DDI coefficients (left) and in the Dicke limit (right) for the same configuration of atoms.

3.5 Coarse-grained approximation

Since we want to describe the system with a large number of atoms, it is advantageous to work out a continuous approximation for our observables. We divide our sample into small slices. Each slice contains N_0 atoms, which are assumed to undergo the same

time evolution [4]. Neglecting the self-action of each slice on itself, we may represent evolution of the whole slice as an evolution of single atom, but with increased number of neighbours. We may effectively increase the number of neighbours by multiplying the term in equations (11)-(12) responsible for interaction between slices by the number of atoms in a slice. We generate an extended pencil-shaped geometry with $L = 15\lambda_0$ and $r = 0.75\lambda_0$. The results for $N = 500$ "clusters" with $N_0 = 10$ atoms in each cluster are shown on fig. 2.

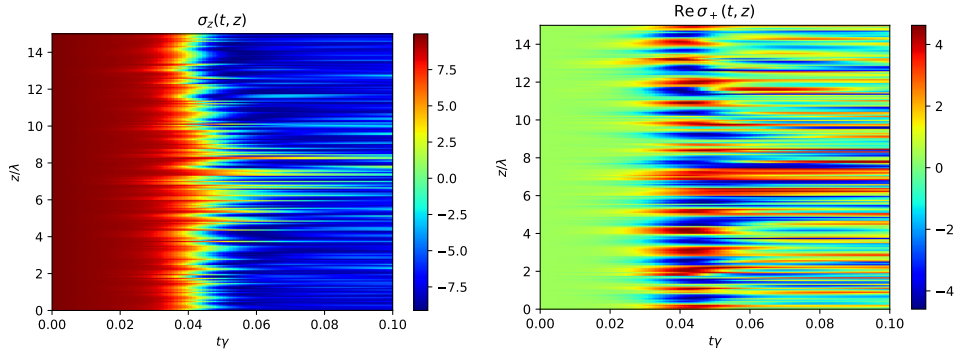

Figure 2: Atomic population $\langle\sigma_z(t,z)\rangle$ on the left and dipole moment $\mathrm{Re}\langle\sigma_+(t,z)\rangle$ on the right for $N = 500$ and $N_0 = 10$ in the complex DDI couplings regime.

The exact solution for $N = 5000$ atoms is shown on fig. 3 (please note that time scale is different). One can see that both systems undergo the same evolution, which indicates the validity of coarse-grained description.

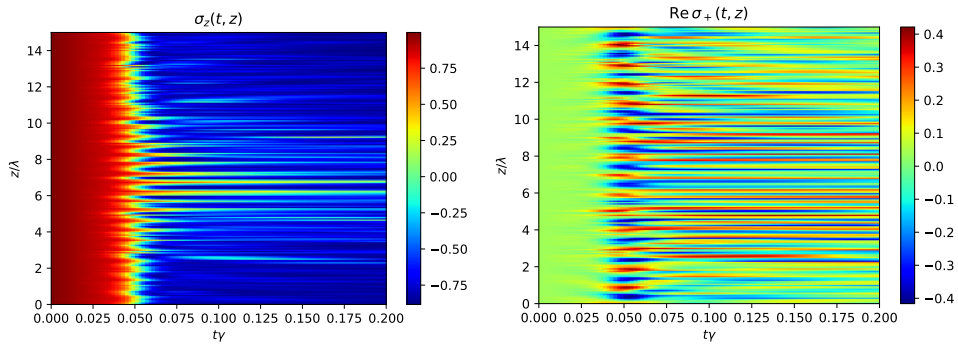

Figure 3: Atomic population $\langle\sigma_z(t,z)\rangle$ on the left and dipole moment $\mathrm{Re}\langle\sigma_+(t,z)\rangle$ on the right for $N = 5000$ and $N_0 = 1$ in the complex DDI couplings regime.

Also note that for such system superradiance occurs in the direction along the atomic sample. This holds for both complex DDI couplings regime and the Dicke limit, which is illustrated on fig. 4. Once again, it indicates the absence of dephasing caused by elastic DDI coefficients.

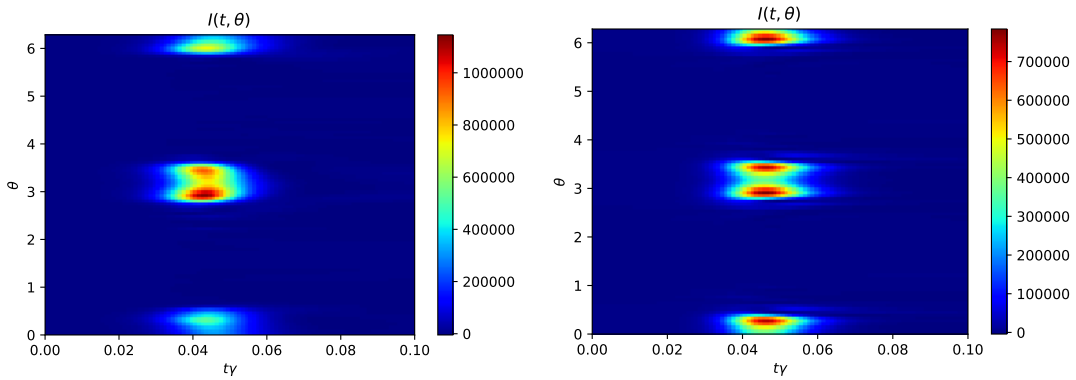

Figure 4: Dependence of intensity of emission $I(t, \theta)$ on the angle θ between the wave-vector direction and z-axis, in a plane formed by z-axis and polarization vector, for complex DDI coefficients (left) and Dicke limit (right).

4 Conslusions

To sum up, in this work we have considered different regimes of superradiance. First, we considered a model with off-resonant terms, which are ommited in the Dicke limit of superradiance. Then we considered a models with forth and back radiation only. In the regime with the complex DDI coefficients, where elastic part is included, the numerical simulation for dense system showed the superradiant behaviour. Also for small samples we have obtained ringings of intensity, which were experimentally observed and which are not present in the Dicke limit. Furthermore, it turns out that there is no dephasing in extended systems as well, and for some set of parameters we have obtained a pronounced propagation along atomic sample. One of the main results of our work is validity of coarse-grained description for extended systems in regime with complex DDI couplings. It opens up many possibilities for probing the parameter values in order to find new effects, as well as to clarify already known results.

References

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