Interaction of a classical laser field with a model Rydberg atom in a mixed state prepared by entanglement with few-photon quantum light

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Abstract: The dynamics a model Rydberg atom prepared initially in a mixed state is investigated in a classical laser field. The possibility of interference stabilization regime is studied and the influence of parameters of initial mixed state on the ionization process is analyzed. In the case when initial atomic mixed state is formed due to entanglement with few-photon non-classical light the possibility to extract the information about initial atomic density matrix from the atomic behavior in the further applied classical laser field is discussed.

Introduction

Today one of the most interesting directions of research in laser physics is the interaction of an atom or molecule with strong electromagnetic field. In the case of strong field new phenomena appear and one of them is stabilization against ionization. Two different mechanisms of stabilization are known: the interference stabilization of Rydberg atoms and the Kramers-Henneberger stabilization. The mechanism of interference stabilization was firstly predicted theoretically for Rydberg atoms interacting with classical laser field [1-5].

However today it is possible to generate different types of non-classical field: squeezed vacuum field [6, 7] and few photon states [8, 9]. That’s why the question about interference stabilization in quantum electromagnetic field becomes relevant. The possibility of interference stabilization of a model Rydberg atom in quantum field was firstly discussed in [10, 11] and the suppression of ionization was established. In addition, an efficient entanglement between atomic and field subsystems was found which provided the atomic system to occupy a certain mixed state due to the interaction with quantum field. Therefore any further dynamics of such an atom should be described in the density matrix formalism. In connection with, two interesting questions appear. At first, what is further dynamics of such specially prepared in a mixed state atom in classical laser field? This is an interesting problem, since it is known that interference stabilization is very sensitive to the phase difference between Rydberg states. But in situation when atom is described by density matrix, such phases can hardly be introduced. The second question concerns the possibility to extract information about the initial density matrix of the atom from its dynamics in a further applied laser field. Such measurements allow to calculate the degree of entanglement [12, 13] induced during the interaction with quantum field [11].
So, in this work the dynamics of a model Rydberg atom prepared initially in a mixed state is considered in classical laser field. The atom is studied in a model which includes several discrete levels and continuum and allows one to investigate both: the field-induced ionization and resonance transitions between low-lying and high exciting atomic states. The possibility of interference stabilization regime in the case of interaction of such atom prepared initially in a mixed state with laser field and the influence of non-diagonal elements of initial density matrix on this dynamics is studied. The possibility to extract the information about initial atomic density matrix from the measurements of ionization and population trapping induced by the classical laser field is discussed.

**Analytical model**

The interaction between an atom and classical field in the density matrix formalism is described by non-stationary equation:

\[
\hbar \frac{\partial \hat{\rho}}{\partial t} = \left[ \hat{H}_{at} + \hat{V}_{\text{int}}, \hat{\rho} \right]
\]

where \( \hat{H}_{at} \) is the atomic Hamiltonian, \( \hat{V}_{\text{int}} = -\vec{d}\vec{E}_0 \cos(\omega t) \) is the atomic-laser field interaction in the dipole approximation, \( \vec{d} \) stands for the dipole moment of atomic electron, and \( \vec{E}_0 \) represents the strength of classical laser field.

In our case the Rydberg atom is considered in a model of “3 bound levels and continuum” and is characterized by 2 highly excited Rydberg levels repopulated by the field-induced \( \Lambda \)-type transitions via the continuum and the resonantly coupled third low-lying bound atomic state. In this case \( \nu \)-type transitions become also possible. The atomic eigenstates obey the stationary Schroedinger equation:

\[
\hat{H}_{at}\varphi_n = E_n \varphi_n \quad n = 0,1,2
\]

\[
\hat{H}_{at}\varphi_E = E \varphi_E
\]

with \( \varphi_n, E_n \) and \( \varphi_E, E \) representing the wavefunctions and eigenvalues of atomic bound and continuum eigenstates respectively.

The frequency of laser field is chosen to provide the exact resonance condition between the lower state and the middle between the two Rydberg levels. We assume that transitions between low-lying atomic state and continuum are negligible small. The considered atomic system is discussed in details in [11]. We considered the hydrogen – like ion with nuclear charge \( Z \), in which fine doublet \( n P_{1/3} \left( \varphi_1, \varphi_2 \right) \) was chosen for highly excited neighboring Rydberg levels and the state \( n_0S \left( \varphi_{0} \right) \) - for the lower lying resonant level. The laser frequency is much larger than the splitting energy \( \Delta = E_2 - E_1 \) between neighboring Rydberg levels and was chosen to correspond to \( \hbar \omega \approx 2eV \) for \( n_0 = 4, Z = 2 \).

Initially the atom is supposed to occupy a certain mixed state which is formed during primary interaction with non-classical few photons field [11]. However, in general, an arbitrary mixed state can be considered as an initial one. The relaxation processes are not taken into account in our case since their characteristic time is of the order of \( 10^{-9} \) s or even more that is much larger than the largest time characterizing the atomic dynamics in a laser field.
Using the mentioned above basis of discrete and continuum atomic states and carrying out the procedure of adiabatic elimination of the continuum [2] we obtain the system of differential equations for density matrix elements between bound atomic states:

\[
\frac{d\rho_{01}}{dt} = \frac{\Delta}{2}\rho_{01} + \Omega(\rho_{11} + \rho_{21} - \rho_{00}) - i\frac{\Gamma}{2}(\rho_{01} + \rho_{02})
\]

\[
\frac{d\rho_{02}}{dt} = -\frac{\Delta}{2}\rho_{02} + \Omega(\rho_{12} + \rho_{22} - \rho_{00}) - i\frac{\Gamma}{2}(\rho_{01} + \rho_{02})
\]

\[
\frac{d\rho_{00}}{dt} = \Omega(\rho_{10} + \rho_{20} - \rho_{01} - \rho_{02})
\]

\[
\frac{d\rho_{11}}{dt} = \Omega(\rho_{01} - \rho_{10}) - i\Gamma\left(\frac{\rho_{12} + \rho_{21}}{2} + \rho_{11}\right)
\]

\[
\frac{d\rho_{12}}{dt} = \Omega(\rho_{02} - \rho_{20}) - i\Gamma\left(\frac{\rho_{12} + \rho_{21}}{2} + \rho_{22}\right)
\]

\[
\frac{d\rho_{21}}{dt} = -\Delta\rho_{12} + \Omega(\rho_{02} - \rho_{10}) - i\Gamma\left(\frac{\rho_{12} + \rho_{21}}{2} + \rho_{12}\right)
\]

where \(\hbar\omega = \frac{E_1 + E_2}{2} - E_0\), \(\Delta = E_2 - E_1\), \(E_0, E_1, E_2\) are the energies of atomic bound states, \(\Omega = \frac{d_{mn}E_0}{2\hbar}\) is the Rabi frequency of resonant transition and \(\Gamma_{mn} = \frac{2n(q^2)}{\hbar}\) are the elements of ionization rate tensor, calculated by using the Fermi golden rule and supposed to be the same and equal to \(\Gamma\).

Because of cumbrous analytical solution the system (2) is solved numerically. The initial density matrix is chosen as that formed during atom-quantum field interaction [11] or in an arbitrary form. The developed approach is rather general and includes the cases of both pure and mixed initial state of the atom. The solution of the system (2) gives the time-dependent population of bound Rydberg states:

\[
W_1(t) = \rho_{11}(t), \quad W_2(t) = \rho_{22}(t), \quad W_0(t) = \rho_{00}(t)
\]

and the probability of ionization of the system:

\[
W_i = 1 - (\rho_{00} + \rho_{11} + \rho_{22})
\]

To analyze the change of initially mixed atomic state during the interaction with laser field we introduce the parameter “purity” \(P\) which is quantitative degree of purity of atomic state and is equal to the trace of atomic density matrix squared.

\[
P = Tr[\rho^2]
\]

Since it is well known that for pure state \(\rho = \rho^2\) and for any density matrix \(\rho \quad Tr[\rho] = 1\), the case \(P = 1\) corresponds to the pure atomic state which can be characterized by a wavefunction and \(P < 1\) – to a mixed state of the atom. Thus, the closer is \(P\) to unity, the closer is the atomic state to the pure state. Since the Eqs. (2) assume possible decay of density matrix elements with time due to atomic ionization the procedure of renormalization is used to obtain the value of parameter \(P\) in a correct way. Finally, it is rewritten as follows:

\[
P(t) = \frac{\rho_{00}^2 + \rho_{11}^2 + \rho_{22}^2 + 2\rho_{12}\rho_{21} + 2\rho_{10}\rho_{01} + 2\rho_{20}\rho_{02}}{(\rho_{00} + \rho_{11} + \rho_{22})^2}
\]

It should be mentioned, that the worked out approach based on the density matrix formalism is a general description of the interaction of a model Rydberg atom with a classical laser field and is valid for both the pure and mixed initial state of the atom. In the case of initial density matrix corresponding to a pure atomic state we immediately obtain from (2) the same results as have been obtained earlier in [3] using the wave function formalism.
Results and discussions

First we investigate the interaction of a model Rydberg atom with a classical laser field in the case when atom is initially in an arbitrary mixed state. Similarly to [3] we consider the regime of rather strong field when the Rabi frequency of resonant transition $\Omega$ is the largest energy parameter in the system and the relations $\Omega \gg \Delta$, $\Omega \gg \Gamma$ are fulfilled. The main feature of the field-induced atomic dynamics is found to consist in the transformation of initial mixed state of the atom into the pure state due to the atom-field interaction during rather short time interval. The typical dependencies of the purity parameter and ionization probability on time are presented on Fig. 1 a, b respectively. The calculations are performed for the following initial density matrix chosen with arbitrary non-zero matrix elements:

$$
\rho_{t=0} = \begin{pmatrix}
0.5 & 0.2 - 0.1 i & 0.1 + 0.3 i \\
0.2 + 0.1 i & 0.2 & 0.1 - 0.2 i \\
0.1 - 0.3 i & 0.1 + 0.2 i & 0.3
\end{pmatrix}
$$

(7)

It is clearly seen that during the atom-field interaction the value of “purity” reaches rather fast its maximal value equal to unity. Thus, the coherent laser field converts the initial atomic mixed state into a pure state inducing certain phases between wave functions in the coherent superposition of bound Rydberg states formed during the interaction. The observed “passage” from initial mixed state to the pure one is accompanied by rather efficient atomic ionization, which, however, saturates at the level significantly less than unity. This means that the field-induced ionization process leads to the “purification” of the bound atomic fraction in such a way that the formed coherent wave packet is characterized by a high resistance to the further ionization. That’s why the system is not fully ionized even in the limit of large time and the probability of ionization becomes constant and doesn’t reach unity. Thus the regime of interference stabilization takes place. It should be emphasized that interference stabilization is known to result from the certain phase relations between bound Rydberg states, while in the case of mixed states the phase information is lost. It is the coherent laser field that brings coherence into incoherent state and induces proper phase relations.

Fig. 1. Parameter “purity” (a) and probability of ionization of the system (b) versus time in optical cycles.

It is rather easy to find that in the steady state regime the residual bound atomic fraction is characterized by the following stable quasienergy wave function:

$$
\psi_{n0}^{QES} = \frac{1}{\sqrt{1 + \frac{\Delta^2}{8\Omega^2}}} \left( \frac{\varphi_1 - \varphi_2}{\sqrt{2}} + \frac{\Delta}{\sqrt{8\Omega}} \varphi_0 \right)
$$

(8)
The expression (8) coincides exactly with the quasienergy state which can be found for the interaction of a laser field with pure state of the atom and also can be obtained from the Schrödinger formalism [3] under proper resonant condition and only one low-lying Rydberg state taken into consideration.

Thus the general feature of the atomic dynamics in a classical laser field is the formation of a pure atomic state which is stable quasienergy state of the system. This effect is general and is observed for arbitrary initial mixed states including different initial pure states as well as thermodynamic mixture with all non-diagonal matrix elements equal to zero. The latest are interesting since all the phase information is absent in such states.

From Eqs. (2) the connection between the ionization probability and the matrix elements of initial density matrix can be found:

$$ W_{\text{ion}} = 1 - W_b = 1 - \rho_{00}|_{t\to\infty} - \rho_{11}|_{t\to\infty} - \rho_{22}|_{t\to\infty} = $$

$$ = 1 - \frac{1}{1 + \frac{\Omega^2}{8\Delta^2}} \left[ \Delta^2 \rho_{00}|_{t=0} + \frac{\rho_{11}|_{t=0} + \rho_{22}|_{t=0}}{2} - \Delta \rho_{12}|_{t=0} + \Delta \rho_{20}|_{t=0} \right] $$

(9)

For example, in the case of the data presented on Fig 1 $\Delta^2 = 1$ and from (9) using the initial condition (7) we obtain $W_{\text{ion}} = 0.6043$. However the inverse problem – to retrieve the information of initial mixed state from ionization dynamics – doesn’t seem to be easy solved, since all elements of initial density matrix should be found while, in fact, only one value - the ionization probability - can be measured.

However in some special cases this problem can be solved. We will show that in the case when the mixed state of the atom is prepared due to its interaction with few-photon quantum light the further applying classical field gives the possibility to measure experimentally the degree of entanglement, which was induced between the atom and quantum light. For this purpose it is necessary to recover the formed density matrix of the atom from its dynamics in classical laser field, because a quantitative degree of entanglement, for example, the value of Schmidt parameter, depends on the components of atomic density matrix. The Schmidt number cannot be obtained by direct experimentally measurement since it depends on both the non-diagonal and diagonal elements of density matrix. The latest are nothing more than a probability of population of Rydberg states and can be in principal measured experimentally.

Further we will demonstrate the possibility to extract the information about the initial atomic density matrix from atomic dynamics in a classical laser field. First, we specify the initial condition in the form of rather general atomic density matrix formed during the interaction of the considered model atom with few-photon quantum field in a steady state regime [11]:

$$ \rho|_{t=0} = \begin{pmatrix} \rho_{00} & 0 & 0 \\ 0 & \rho_{11} & \rho_{21} \\ 0 & \rho_{12} & \rho_{22} \end{pmatrix} $$

(11)

This density matrix can be found from the evolution of the system “atom+ quantum field” which is shown to transfer the system to the following bipartite quasienergy state [11]:

$$ \psi^{\text{QES}} = \left( \alpha \Phi_k - \beta \Phi_{k-1} + \beta \phi_0 \Phi_k \right) $$

(12),

where $\Phi_k$ is k-photon Fock state and the amplitudes $\alpha, \beta$ are determined by the atomic and quantum field parameters and obey the normalization condition: $|\alpha|^2 + |\beta|^2 = 1$. 

The corresponding reduced density matrix describing the atom in a quantum field in a steady state regime is given by:

\[
\rho_{\text{at}}^r = \frac{|\alpha|^2}{2} \begin{pmatrix} |\beta|^2 & 0 & 0 \\ 0 & 1 & -1 \\ 0 & -1 & 1 \end{pmatrix} \tag{13}
\]

It is important that such matrix has zero elements: \(\rho_{10} = \rho_{20} = \rho_{01} = \rho_{02} = 0\) due to the resonant transitions \(\varphi_0 \leftrightarrow \varphi_{1,2}\) providing these states always associated with different number of photons.

It is convenient to move from initial basis of bound atomic states to the new basis which provides the initial density matrix (11) to be diagonal. This basis exactly coincides with Schmidt modes found for atomic subsystem in the case of the steady state (12) describing the entanglement between the atom and few-photon quantum field. By solving the eigenproblem for such reduced atomic density matrix (11) one can obtain the following expression for Schmidt modes or new basis atomic states:

\[
\tilde{\varphi}_0 = \varphi_0, \quad \tilde{\varphi}_1 = \frac{\rho_{21}}{\sqrt{\rho_{12}}}\varphi_1 + \varphi_2, \quad \tilde{\varphi}_2 = \frac{\varphi_1 - \rho_{21} \varphi_2}{\sqrt{2}}\tag{14}
\]

The basis of Schmidt modes is seen to depend strongly on the ratio \(\frac{\rho_{21}}{\rho_{12}}\) and the initial density matrix becomes diagonal in terms of these states instead of being non-diagonal in the previous basis. However, the diagonal elements correspond now to the population of the Schmidt states that hardly can be measured experimentally. What is also very important is that for the quasienergy state (12) \(\rho_{12} = \rho_{21}\) due to the stabilization regime found for the Rydberg atom in few photon quantum field. Under this condition the Schmidt modes do not depend on the amplitudes \(\alpha, \beta\) and are given by:

\[
\tilde{\varphi}_0 = \varphi_0, \quad \tilde{\varphi}_1 = \frac{\varphi_1 + \varphi_2}{\sqrt{2}} \equiv 0, \quad \tilde{\varphi}_2 = \frac{\varphi_1 - \varphi_2}{\sqrt{2}} \tag{15}
\]

And the initial atomic mixed state is characterized by the diagonal matrix written as follows:

\[
\tilde{\rho}_{|t=0} = \begin{pmatrix} \tilde{\rho}_{00}|_{t=0} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \tilde{\rho}_{22}|_{t=0} \end{pmatrix} = \begin{pmatrix} |\beta|^2 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & |\alpha|^2 \end{pmatrix} \tag{16}
\]

Our goal now is to retrieve only one of the two diagonal matrix elements (due to the normalization condition) performing one measurement only. The system (2) can be rewritten in the Schmidt basis (15) by following way:

\[
\frac{i\hbar}{\Delta} \frac{d\tilde{\rho}_{10}}{dt} = -\frac{\Delta}{2} \tilde{\rho}_{20} - \sqrt{2}\Omega (\tilde{\rho}_{11} - \tilde{\rho}_{00}) - i\Gamma \tilde{\rho}_{10}
\]

\[
\frac{i\hbar}{\Delta} \frac{d\tilde{\rho}_{20}}{dt} = -\frac{\Delta}{2} \tilde{\rho}_{10} - \sqrt{2}\Omega \tilde{\rho}_{21}
\]

\[
\frac{i\hbar}{\Delta} \frac{d\tilde{\rho}_{00}}{dt} = \sqrt{2}\Omega (\tilde{\rho}_{10} - \tilde{\rho}_{01})
\]

\[
\frac{i\hbar}{\Delta} \frac{d\tilde{\rho}_{11}}{dt} = -\sqrt{2}\Omega (\tilde{\rho}_{10} - \tilde{\rho}_{01}) - i\Gamma \tilde{\rho}_{11} - \frac{\Delta}{2} (\tilde{\rho}_{21} - \tilde{\rho}_{12}) \tag{17}
\]

\[
\frac{i\hbar}{\Delta} \frac{d\tilde{\rho}_{22}}{dt} = \frac{\Delta}{2} (\tilde{\rho}_{21} - \tilde{\rho}_{12})
\]

\[
\frac{i\hbar}{\Delta} \frac{d\tilde{\rho}_{21}}{dt} = -\sqrt{2}\Omega (\tilde{\rho}_{20} + \tilde{\rho}_{02}) - i\Gamma \tilde{\rho}_{21} - \frac{\Delta}{2} (\tilde{\rho}_{11} - \tilde{\rho}_{22})
\]
It’s easy to see from (17) that some initially zero matrix elements \(\text{Re}[\tilde{\rho}_{10}], \text{Im}[\tilde{\rho}_{20}], \text{Re}[\tilde{\rho}_{21}]\) remain equal to zero during the dynamics of the system. However some of them being initially zero \(\tilde{\rho}_{11}, \text{Im}[\tilde{\rho}_{10}], \text{Im}[\tilde{\rho}_{21}]\) vary during the dynamics of the system but turn to zero again in the steady regime of the interaction. This feature is a direct consequence of the interference stabilization phenomenon and the formation of a stable atomic pure state (8) (from the initial mixed one) in a strong classical laser field. Fig. 2 represents the growth of the population of initially non-populated Schmidt state \(|\tilde{\phi}_{1}\rangle\) due to the interaction with laser field and its decrease up to zero in the steady state regime when the bound atom is characterized by a pure quasienergy state (8) resistant to the further ionization.

\[
\text{Fig 2. Dynamics of population of initially non-populated Schmidt state } |\tilde{\phi}_{1}\rangle \text{ versus time in optical cycles.}
\]

In terms of Schmidt basis the steady density matrix corresponding to the stable atomic state (8) is given by:

\[
\tilde{\rho}|_{t\to\infty} = \frac{W_b}{1 + \frac{\Delta^2}{8\Omega^2} + \frac{\Delta}{\sqrt{8\Omega}}} \begin{pmatrix}
\frac{\Delta^2}{8\Omega^2} & 0 & \frac{\Delta}{\sqrt{8\Omega}} \\
0 & 0 & 0 \\
\frac{\Delta}{\sqrt{8\Omega}} & 0 & 1
\end{pmatrix},
\]

(18),

where \(W_b\) denotes the residual bound probability of the atom. Notice, the fact that \(\text{Tr}[\tilde{\rho}|_{t\to\infty}] = W_b < 1\) is a consequence of the decay of the atomic bound fraction due to the ionization process. Thus the evolution of atomic system in a laser field starting from a mixed state (16) comes to the pure state (18).

It should be emphasized that from the system of differential equations in Schmidt basis (17) a certain combination of density matrix elements \(S\) is found to be conserved during the interaction with the laser field and its derivative is identically zero at any instants of time:

\[
\imath\hbar \frac{d}{dt} \left[ \frac{\Delta^2}{8\Omega^2} \tilde{\rho}_{00} + \tilde{\rho}_{22} + \frac{\Delta}{\sqrt{8\Omega}} \text{Re}[\tilde{\rho}_{20}] \right] \equiv \imath\hbar \frac{d}{dt} S = 0
\]

(19)

It means that such value is directly proportional to the residual bound probability surviving in a classical laser field. Indeed, from (18) we immediately obtain:

\[
S = W_b(1 + \frac{\Delta^2}{8\Omega^2})
\]

(20)

From the other hand, the value of \(S\) can be obtained from the initially density matrix (16). Equating both values we obtain:

\[
\frac{\Delta^2}{8\Omega^2} \tilde{\rho}_{00}|_{t=0} + \tilde{\rho}_{22}|_{t=0} + \frac{\Delta}{\sqrt{8\Omega}} \text{Re}[\tilde{\rho}_{20}|_{t=0}] = W_b(1 + \frac{\Delta^2}{8\Omega^2})
\]

(21)

In account of the zero values of the non-diagonal element \(\tilde{\rho}_{20}|_{t=0}\) we obtain the connection between the non-zero matrix elements of initial density matrix (16) describing the initial atomic mixed state and the final probability of ionization of the atom in a laser field:

\[
W_{ion} = 1 - \rho_{00}|_{t\to\infty} - \rho_{11}|_{t\to\infty} - \rho_{22}|_{t\to\infty} = 1 - \frac{1}{1 + \frac{\Delta^2}{8\Omega^2}} \left[ \frac{\Delta^2}{8\Omega^2} \tilde{\rho}_{00}|_{t=0} + \tilde{\rho}_{22}|_{t=0} \right]
\]

(22)
where the diagonal elements are connected with each other by the condition of the unity trace.

The obtained expression (22) means that by measuring the probability of ionization in the steady state regime during the interaction of a Rydberg atom prepared initially in a mixed state with classical electromagnetic field we can retrieve the initial atomic density matrix in the Schmidt basis and therefore calculate the degree of entanglement (the value of the Schmidt parameter $K$) between the atom and few-photon non-classical light induced during their interaction before the classical field is applied:

$$ K^{-1} = \rho_{00} |_{t=0}^2 + \rho_{22} |_{t=0}^2 = P |_{t=0} $$  \hspace{1cm} (23)

Thus it is possible to measure the degree of entanglement between the atom and few-photon quantum light by further applying of classical laser field.

**Conclusion**

The interaction of a model Rydberg atom prepared initially in a mixed state with classical electromagnetic laser field is studied. A general feature of atomic dynamics is found to consist in the transformation of the initial mixed state of the atom to the pure state due to the interaction with laser field. In other words, classical field brings coherence into initially incoherent state. The “purification” is accompanied by significant ionization of the atom. However the laser field induces certain phases between bound Rydberg states composing the formed coherent wave packet in such a way that the residual atomic population becomes resistant to further ionization. In the steady state regime the regime of interference stabilization takes place and the bound atomic fraction is described by the stable quasienergy state of the system. The observed “purification” is a common feature of atomic dynamics in a classical laser field and can be used as a tool to retrieve the information about initial density matrix of the atom. In the case when the initial mixed state of the atom is formed during its primary interaction with few-photon quantum field it is possible to extract the information about the mixed atomic state and therefore about the entanglement reached between the atom and few-photon field by performing only one measurement of the probability of ionization induced by applying classical laser light.

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