Enhanced nonlinearity of four-wave mixing via Rydberg–Rydberg interactions

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Abstract

We investigate the four-wave mixing (FWM) nonlinearity in an ensemble of cold Rydberg atoms with each of them regarded as a double-ladder system. The interaction is studied from the view of generating a signal field in virtue of three applied lasers. Using an approach beyond mean-field theory, we solved the equations for the one-body and two-body correlators under perturbation, and show that the system possesses not only a local FWM nonlinearity, but also a much larger nonlocal nonlinearity due to the Rydberg–Rydberg interaction which can be further strengthened by increasing the atomic density. The results obtained may have promising applications in the quantum information processes involving the FWM nonlinearity, such as the generation of squeezed or entangled states.

1. Introduction

Atoms excited to high-lying Rydberg states interact via strong and long-range dipole–dipole or van der Waals forces, and such strong interactions manifest as a well-known excitation blockade [1] in which an atom promoted to a Rydberg level shifts the energy levels of nearby atoms and suppresses their excitation. The blockade is of great interest for applications in quantum information [2] and provides the basis for numerous proposals, such as the photon–photon phase gates [3], atomic logical gates [4, 5], single-photon switch [6] and generating nonclassical state of light [7, 8].

Due to such pronounced nonlocal interaction between the atoms, the Rydberg atomic gases are regarded as a promising candidate for many-body correlated systems which act as effective tools to control the interactions between photons [9, 10]. One of such applications is to enhance nonlinearity [11–13] with the help of electromagnetically induced transparency (EIT). For example, the Kerr effect, as one of main topics in nonlinear optics with numerous applications in, such as single-photon switches and transistors [9], controlled quantum gates [14–16], quantum teleportation [17] and entanglement [18], can be greatly enhanced in Rydberg-EIT system [19–21]. Calculations beyond mean-field theory [21–25] attribute such improvement to the two-body or even three-body correlators based on perturbation method.

Motivated by the recent investigations on strengthening the Kerr nonlinearity, in this paper we study the enhancement of another third-order nonlinearity, namely, the four-wave mixing (FWM) process which has been used as an important resource for many quantum applications, such as quantum entanglement and steering [26–30], quantum cloning [31], generating correlated beams and photon pairs [32–35], constructing nonlinear interferometer [36] and quantum networks [37], as well as realizing optical nonreciprocity [38], etc.

The system that we are interested in is shown in figure 1(a) where three applied fields, specifically, a pumping field (\(\Omega_p\)), coupling field (\(\Omega_c\)) and a driving field (\(\Omega_d\)) illuminate the four-level atoms with a Rydberg state as the highest level to generate a signal (\(\Omega_s\)) with the efficiency determined by the third-order susceptibility. Investigations using the similar excitation schemes are reported, for example, to build a single-photon source by utilizing the Rydberg blockade of confined atoms in an excitation volume similar...
to, or smaller than that of the blockade sphere [39–41], or using the pumping and coupling fields to construct Rydberg dark-state polaritons for, e.g., the efficient light storage which can be retrieved by applying the driving field after a short time period [42, 43]. We also note the early experimental investigations on FWM in Rydberg gases of which the atomic density is too low to invoke significant Rydberg–Rydberg interaction [44–47]. In this paper, we study instead a (virtual) transition between the Rydberg atom at position \( \mathbf{r} \) and the one at origin) and three-atom correlation (including one more atom at \( \mathbf{r}' \)) are discussed in this paper.

At first glance, the nonlinear susceptibility of the FWM process has a small modulus, meaning that the generation of \( \Omega \) cannot be efficient, as the susceptibility is proportional to the product of the four relevant dipole-moment elements in which the ones associating with the transitions of \( \delta r \) and the photon are interested in this paper.

We assume that the electric fields can be written as \( E_i = \mathbf{\hat{r}}_i e^{i k \mathbf{r} \cdot \mathbf{r} + i \omega_{ni} t} + c.c., \) \( i = \{ p, c, d, s \} \) with c.c. standing for complex conjugate. Then the corresponding Rabi frequencies are \( \Omega_i = \mu_{mn} \mathbf{\hat{r}}_i / \hbar \), with \( m, n = \{ 1, 2, 3, 4 \} \) being the dipole moment of the transition \( |m\rangle \leftrightarrow |n\rangle \) that \( \Omega_i \) drives. And the detunings are defined as \( \Delta_{r} = \omega_{21} - \omega_{p}, \Delta_{c} = \omega_{d4} - \omega_{21} - \omega_{c}, \Delta_{d} = \omega_{21} - \omega_{31} - \omega_{s}, \Delta_{s} = \omega_{31} - \omega_{r} \).

Adopting the similar notations used in reference [24], we use \( \hat{S}_{mn}(r, t) = |m(r, t)\rangle \langle n(r, t)| \) to denote the transition operators for the atom at \( r \) and \( \mathbf{r} \), and it satisfies the commutation relation \( \hat{S}_{ab}(r, t) \), \( \hat{S}_{mn}(r', t) = [\delta_{mn} \hat{S}_{ab}(r, t) - \delta_{ab} \hat{S}_{mn}(r', t)] \delta_{rr} \) where \( \delta_{ab} \) is Kronecker delta symbol. In the derivation to follow, we use \( \hat{S}_{mn}(t) \) to represent the transition operator \( \hat{S}_{mn}(0, t) \) for the atom at origin, then under the

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**Figure 1.** (a) The four-level atomic system with a Rydberg state \( |4\rangle \) illuminated by a pumping field \( \Omega_p \), coupling field \( \Omega_p \) and a driving field \( \Omega_d \) to generate signal field \( \Omega_s \). (b) The coordinate system for calculating the Rydberg–Rydberg interaction, with the atom of interest located at origin. The blocked sphere caused by the interaction between Rydberg atoms (red dots) is schematically shown by the blue dashed circle. Inside each sphere, only one atom can be excited to the Rydberg state while excitations of other atoms (gray dots) are prevented. The correlation between the two Rydberg atoms (e.g. the one at position \( \mathbf{r} \) and the one at origin) and three-atom correlation (including one more atom at \( \mathbf{r}' \) ) are discussed in this paper.
electric-dipole and rotating-wave approximations, the Hamiltonian depicting the interaction between the atom (at origin) and the four fields takes the following form:

\[
\hat{H}(t) = \Delta_C \hat{S}_C(t) + \Delta_p \hat{S}_p(t) + \Delta_t \hat{S}_t(t)
\]

Here \(N_0\) is the atomic density and the symbol \(\int d^3r\) stands for the integration \(\int_0^\pi \sin \theta d\theta \int_0^{2\pi} d\phi \int_{R_0}^{\infty} r^2 dr\) in spherical coordinates with \(R_0\) being the radius of the blockage sphere (see figure 1(b)). Then the features of the generation, absorption, as well as dispersion of the signal \(\Omega\), all hide in \(\langle \hat{S}_3(\tau) \rangle = \rho_{33}(t)\) which is one of the one-body density-matrix elements governed by \(i\hbar \partial \langle \hat{S}_{3n}(t) \rangle / \partial t = \langle [\hat{S}_{3n}(t), \hat{H}] \rangle\). For instance, the equation of \(\rho_{33}(t)\) reads

\[
\frac{\partial \rho_{33}}{\partial t} = -g_{33} \rho_{33} + i\Omega_3 (\rho_{11} - \rho_{33}) - i\rho_{32} \Omega_p + i\rho_{41} \Omega_4^*;
\]

(2a)

it further relates to other elements, e.g. \(\rho_{41}\) whose time dependence is:

\[
\frac{\partial \rho_{41}}{\partial t} = -g_{41} \rho_{41} + i\rho_{21} \Omega_c + i\rho_{31} \Omega_d - i\rho_{42} \Omega_p
\]

\[
- i\rho_{43} \Omega_s - iN_0 \int d^3r V(r) \rho_{44,41}(r, t).
\]

(2b)

Here \(g_{33} = \gamma_{31} + i\Delta_p\) and \(g_{41} = \gamma_{41} + i(\Delta_c + i\Delta_p)\). The whole (closed) set of the equations for the one-body density-matrix element is listed in the appendix A. The last term on the right-hand side of equation (2b) comes from the Rydberg–Rydberg interaction where \(\rho_{44,41} = \langle \hat{S}_{44}(r, t) \hat{S}_{31}(t) \rangle\) is one of the two-body correlators, generally defined as \(\rho_{ab,mn}(r, r', t) = \langle \hat{S}_{ab}(r, t) \hat{S}_{mn}(r', t) \rangle\). Clearly, to solve the equations of the one-body density-matrix elements we need to find the values of the two-body correlators first. Akin to these one-body objects, the two-body elements satisfy a new set of the equations. For example, the equation for \(\rho_{44,41}\) is

\[
\frac{\partial \rho_{44,41}}{\partial t} = -\left[g_{41} + \Gamma_{42} + \Gamma_{43} + iV(r)\right] \rho_{44,41}
\]

\[
- i\Omega_4^* \rho_{42,41} - i\Omega_3 \rho_{43,41} + i\Omega_c (\rho_{44,21} + \rho_{41,24})
\]

\[
+ i\Omega_d (\rho_{44,31} + \rho_{41,34}) - i\Omega_p \rho_{44,42} - i\rho_{44,43}
\]

\[
- iN_0 \int d^3r V(r) \langle \hat{S}_{44}(r', t) \hat{S}_{44}(r, t) \hat{S}_{31}(t) \rangle.
\]

(3a)

It depends on other two-body correlators, we present in the following the equations for only two of them, so as to show the key features and save space in the meantime.

\[
\frac{\partial \rho_{44,21}}{\partial t} = -(g_{21} + \Gamma_{22} + \Gamma_{23} + iV(r)) \rho_{44,21}
\]

\[
+ i\Omega_c \rho_{24,21} - i\Omega_3 \rho_{23,21} - i\Omega_p \rho_{44,21}
\]

\[
- i\rho_{43,21} - i\rho_{44,23} - i\rho_{42,23},
\]

(3b)

\[
\frac{\partial \rho_{41,24}}{\partial t} = -(g_{24} - g_{41}) \rho_{41,24} + i\Omega_c \rho_{24,21}
\]

\[
- i\Omega_3 (\rho_{41,22} - \rho_{44,41}) + i\Omega_p (\rho_{44,41} - \rho_{42,24}) - i\rho_{43,24}
\]

\[
- iN_0 \int d^3r V(r) \langle \hat{S}_{44}(r, t) \hat{S}_{41}(t) \hat{S}_{24}(r', t) \rangle
\]

\[
+ iN_0 \int d^3r V(r) \langle \hat{S}_{41}(r', t) \hat{S}_{44}(r, t) \hat{S}_{24}(t) \rangle.
\]

(3c)
Figure 2. Hierarchy of perturbation. The original system (a) is expanded with respect to the signal field (column b). We only consider the (0)-order system in panel (b1) here. The (1)-order system in panel (b2), and the following other-order systems related to Raman enhancement, Kerr nonlinearity and more higher-order effects experienced by the signal are out of the scope of this article. The (0)-order system is further expanded with respect to the pumping field (column c). As we can see that the order of perturbation is represented by the number of arrows of $\Omega_p$. The two-body correlator that interests us ($\rho_{\langle 3 \rangle 44,41}$) seats on the $\langle 3 \rangle$ order. And the corresponding panel follows next to (c3), however, is not explicitly shown.

As we can see that the above equations contains three-body correlators, such as $\langle \hat{S}_{44}(r', t)\hat{S}_{44}(r, t)\hat{S}_{41}(t) \rangle$. One can easily foresee that the motion of equations for the three-body correlators depend on the four-body ones and so on.

3. The local and nonlocal nonlinear susceptibilities

To achieve an effective FWM interaction, the excitation scheme illustrated in figure 1(a) uses a far-off-resonance pumping field $\Omega_p$ to avoid the excitation of the atoms to level $|2\rangle$. In virtual of the strong coupling field, a dark state [49] can be formed between $|1\rangle$ and $|4\rangle$ which further reduces the population on $|2\rangle$. Further considering that the Rydberg–Rydberg interaction causes energy shift which leads the blockade effect, then we can conclude that the population on $|4\rangle$ is nearly negligible. Under the driving field $\Omega_d$, the FWM process, that is the virtual transition $|1\rangle \rightarrow |2\rangle \rightarrow |4\rangle \rightarrow |3\rangle \rightarrow |1\rangle$ is initiated and results in the generation of $\Omega_s$ without the other transition processes drawing energy from the inputs.

From this point of view, this double-ladder system is very similar to the double-$\Lambda$ system that has been used in generating the entangled photon pairs [50, 51]. The difference is that the level $|4\rangle$ is one of the ground levels in the double-$\Lambda$ configuration, instead of a high excited Rydberg state. And we show in the following that this makes a big difference in the underlying physics.

The arrangement of the applied fields allows us to solve the equations of the density-matrix elements using the perturbation method with respect to the far-detuned pumping field and generated (weak) signal field. And the procedure is shown in figure 2 with panel (a) representing our original system. First, we expand the one-body elements with respect to $\Omega_p$ as

$$\rho_{mn} = \rho_{mn}^{(0)} + \rho_{mn}^{(1)} + \rho_{mn}^{(2)} + \cdots.$$  \hspace{1cm} (4a)

And the (0)-order equations obtained from the perturbation is simply the equations (A1)–(A7) listed in appendix A after setting $\Omega_s = 0$. The interaction corresponding to the (0)-order equations is shown in panel (b1) of figure 2, as we can see the signal field is not included. Under the influences of the pumping, coupling and driving fields, a polarization proportional to $\rho_{11}^{(0)}$ is built between $|1\rangle$ and $|3\rangle$, and illustrated vividly in panel (b1) that $|1\rangle$ is connected with $|3\rangle$ by the three applied fields. Naturally, the polarization takes a form of $\mathcal{P}_s = \chi^{(3)} E_p E_c E_d$ and leads to the generation of the signal depicted by the effective Hamiltonian [52, 53] that $\propto \chi^{(3)} E_p E_c E_d$ with $\hat{E}_s$ associated with the creation operator at $\omega_s$. We shall not further discuss in detail about the generation but only focus on coefficient $\chi^{(3)}$. And the first-order
system shown in (b2) is not discussed either in this paper, as it relates to the absorption and dispersion of the signal field after being generated, or of a field that is deliberately applied. The solution of \( \rho_{31}^{(0)} \) provides the exact result of the nonlinear coefficient. However the result is still quite complex. Fortunately, the ladder-type EIT system (\( |1\rangle - |2\rangle - |4\rangle \)) employs a far-detuned pumping field which naturally becomes our next perturbation parameter with its zeroth-order system shown in panel (c1), the first-order system in panel (c2) and the second in (c3). The corresponding expansion of the elements is

\[
\rho_{31}^{(1)} = \rho_{31}^{(0)} + \rho_{31}^{(1)} + \rho_{31}^{(2)} + \cdots.
\]  

(4b)

Note that the order of perturbation over \( \Omega_p \) is emphasized by angle brackets \( \langle \cdot \rangle \) in the superscripts. The \( \langle 0 \rangle \)-order system in panel (c1) is described by a set of equations with \( \Omega_p = 0 \) and \( \Omega_s = 0 \). Then one can easily see that this is a trivial system. Without the pumping and the signal, the atoms stay on the ground state and the corresponding elements of density matrix are zeros except \( \rho_{11}^{(0)} = 1 \). Then the \( \langle 1 \rangle \)-order system in panel (c2) with the corresponding equations listed in appendix A as equation (A8), can help us to reveal the FWM nonlinearity. Simple calculations lead to

\[
\rho_{31}^{(1)} = -\frac{i}{\epsilon_\alpha} \Omega_c \Omega_d \Omega_p + N_{d,21} \Omega_d \frac{4\pi}{\alpha} \int_0^\infty dr V(r) r^2 \rho_{44,41}(r, t). \tag{5}
\]

Here \( \alpha = g_{c4}(|1\rangle^2 + g_{c4}|3\rangle^2 + g_{d4}|4\rangle^2 \). The polarization built between \( |3\rangle \) and \( |1\rangle \) is formed via two different mechanisms represented by the two terms on the right-hand side of equation (5). An atom acting with the applied field as a independent (isolated) object leads to the generation of \( \Omega_s \) via a local nonlinearity. Considering the definitions of the Rabi frequencies and the polarization \( \mathcal{P}_s = 2N_0 \mu_{31} \rho_{31}^{(1)} \), we find the corresponding coefficient is

\[
\chi_d^{(3)} = -N_0 U_0 \frac{i \mu_{31}^2}{g_{31}(|1\rangle^2 + g_{d4}|4\rangle^2 + g_{c4}|3\rangle^2)). \tag{6}
\]

With

\[
U_0 = \frac{\mu_{21} \mu_{42} \mu_{44} \mu_{31}}{4 \epsilon_\alpha h^3 \omega_{31}}, \tag{7}
\]

And \( N_0 U_0 \) has a dimension of a third-order nonlinear susceptibility. In other words, if the level \( |4\rangle \) is not a Rydberg state, then the atomic gases would provide a nonlinearity whose strength is well modeled by \( \chi_d^{(3)} \). However the level \( |4\rangle \) being a Rydberg state with a extreme long lifetime makes the local FWM process less efficient because \( \mu_{42} \) and \( \mu_{43} \) are much smaller than that of a transition involving only the non-Rydberg states.

The second term in equation (5) comes from the Rydberg-Rydberg interaction and we show in the following that this part of interaction dominates in the FWM process. Note that \( \rho_{44,41}(r, t) \) in equation (5) is a full-order quantity with respect to \( \Omega_p \). We follow a similar procedure as in equation (4) to expand the two-body correlators as

\[
\rho_{ab,mm}^{(0)} = \rho_{ab,mm}^{(0)} + \rho_{ab,mm}^{(1)} + \rho_{ab,mm}^{(2)} + \cdots, \tag{8a}
\]

\[
\rho_{ab,mm}^{(0)} = \rho_{ab,mm}^{(0)} + \rho_{ab,mm}^{(1)} + \rho_{ab,mm}^{(2)} + \cdots. \tag{8b}
\]

The \( \langle 0 \rangle \)-order elements \( \rho_{ab,mm}^{(0)} \) are zeros since the probability of finding an atom on Rydberg state is zero without pumping and signal [see, panel (c1)]. The \( \langle 1 \rangle \)-order elements \( \rho_{ab,mm}^{(1)} \) are zeros as well, due to the fact that the probability of finding a Rydberg atom located at \( r \) is still very small under the large-detuned pumping (at most on the second order of \( \Omega_p \) ) [24].

Calculations on perturbation show that the \( \langle 2 \rangle \)-order non-trivial elements (corresponding to panel (c3)) belongs to two sets of closed equations which are presented in matrix forms as equations (A10) and (A11) in appendix A. The lowest order of two-body correlator \( \rho_{44,41} \) which appears in equation (5) is \( \langle 3 \rangle \), and based on equation (3a), we have

\[
[g_{41} + \Gamma_{42} + \Gamma_{43} + i V(r)] \rho_{44,41}^{(3)} = i \Omega_e \left( \rho_{44,21}^{(3)} + \rho_{41,24}^{(3)} + i \Omega_d \left( \rho_{44,31}^{(3)} + \rho_{41,34}^{(3)} \right) \right) - i \Omega_e^* \rho_{42,41}^{(3)} - i \Omega_d^* \rho_{43,41}^{(3)} \tag{9a}
\]
The above equation depends on other (3)-order two-body elements, such as \( \rho_{44,21}^{(3)} \) and \( \rho_{41,24}^{(3)} \). Based on equations (3b) and (3c), they satisfy

\[
\begin{align*}
(g_{21} + \Gamma_{42} + \Gamma_{43})\rho_{44,21}^{(5)} & = -i\Omega_{4d}\rho_{43,21}^{(3)} + i\Omega_p\rho_{44}^{(2)} \\
& + i\Omega_p\rho_{24,21}^{(3)} - i\Omega_c^*(\rho_{22,21}^{(3)} - \rho_{44,41}^{(3)}) + i\Omega_{4d}\rho_{44,21}^{(3)}; \\
(g_{24} + g_{41})\rho_{41,24}^{(5)} & = i\Omega_{4d}\rho_{31,24}^{(3)} - i\Omega_c^*\rho_{41,23}^{(3)} \\
& + i\Omega_{4d}\rho_{41,14}^{(3)} + i\Omega_p\rho_{41,24}^{(2)} - i\Omega_c^*(\rho_{41,22}^{(3)} - \rho_{44,41}^{(3)}).
\end{align*}
\]

The whole set of the algebraic equations for the (3)-order two-body elements has 27 unknowns including \( \rho_{44,41}^{(3)}, \rho_{43,21}^{(3)} \) and \( \rho_{41,24}^{(3)} \) which are listed in appendix A as an array, see equation (A12). As for their algebraic equations, we find that they are too tedious and not suitable to be presented in this paper even for appendix A, thus we only provide equation (9) here as examples. Note that those (3)-order equations relate to the (2)-order one/two-body elements. And the (2)-order elements depend on (1)-order elements whose equations can be easily solved. Unfortunately, the solution to \( \rho_{44,41}^{(3)} \) does not have a compact form, but we managed to write the expression into a power series with respect to \( \Omega_c \) and \( \Omega_d \):

\[
\rho_{44,41}^{(3)}(r) = \sigma(r)\Omega_c\Omega_d.
\]

With

\[
\begin{align*}
\sigma(r) & = -|\Omega_p|^2\left\{ |\Omega_c|^2 \right. \\
& \times \frac{1}{2\beta(r)\gamma_{41}(g_{24} + g_{41})} \left[ \frac{g_{31}}{\alpha(g_{41} + g_{12})} + \text{c.c.} \right] \\
& + \frac{\beta(r)(\Gamma_{42} + \Gamma_{43})/(g_{21} + \Gamma_{42} + \Gamma_{43})}{\alpha g_{42}} \left[ \frac{g_{31}}{\alpha g_{42}} + \text{c.c.} \right] \\
& \left. + \frac{2g_{31}}{\alpha\beta(r)(g_{21} + g_{41})(2g_{41} + iV(r))\left[g_{41} + g_{42} + iv(r)\right]} + \cdots + Q(m, n)|\Omega_c|^{2m}|\Omega_d|^{2n} + \cdots \right\}.
\end{align*}
\]

With \( m = 2, 3, \ldots \), and \( n = 1, 2, \ldots \). The parameter \( \alpha \) is defined in the previous discussion and \( \beta(r) = g_{41} + \Gamma_{42} + \Gamma_{43} + iV(r) \). \( Q(m, n) \) represents a general coefficient of the series. The exact solution of \( \rho_{44,41}^{(3)} \) should include all terms of the above expansion, and we have to calculate it numerically. From the equation (11) we can see that the resultant nonlinear coefficient depends on the squared modular of the coupling and driving Rabi frequencies, suggesting that nonlocal FWM process is dressed by the strong fields.

Substituting equations (10) and (11) into equation (5), we find nonlocal nonlinear coefficients is

\[
\chi_R^{(3)} = \frac{N^2_0U_0^7\gamma_3^4\pi^4}{\alpha} \int_{0}^{\infty} drV(r)r^2 \sigma(r).
\]

In evaluating (12), we first determine the value of \( R_0 \), which under large pumping detuning, is \( R_0 = \sqrt{\gamma_3/\theta_{\text{ETT}}} \) with \( \theta_{\text{ETT}} = \Omega_c^2/\Delta_p \) \([13, 54]\), then the result is obtained from the integral with simple calculations, and is shown in figure (3).

Based on the results of \( \Re \chi_R^{(3)} \) in figure 3(a) and \( \Im \chi_R^{(3)} \) in (b), we see that the nonlocal nonlinearity reaches its maximal strength at the double-photon resonance \( \Delta_c + \Delta_p = 0 \). This is physically intuitive as, under the same condition, the maximal coherence between \( |1 \rangle \) and \( |4 \rangle \) is achieved. The center frequency of the generated field must satisfy \( \Delta_p + \Delta_c = \Delta_4 + \Delta_\chi \) due to the conservation law of the energy. And for the parameters we used in figure 3, \( \Delta_\chi = 50\gamma_3 \).

As comparison we plot the local nonlinear susceptibilities in figure 3 as well to show that amplitude of \( \chi_R^{(3)} \) is much larger than that of \( \chi_a^{(3)} \) at the given atomic density. The local nonlinearity comes solely from the interaction between the photon and the atoms which can be viewed, as we stated before, a coherence between \( |1 \rangle \) and \( |3 \rangle \) built by the three applied fields leading to a polarization that induces the emission of the signal.

On the other hand, the nonlocal nonlinearity has a different mechanism. In virtue of the coupling and pumping field, a few atoms are excited to the Rydberg states and repel each other via the extra large dipole moments. This causes the addition potential energy \( V(r) \) which at the same time suggesting the existence of a tendency that the atomic gases would reduce its energy to a low state. Such tendency corresponds to the two-body element in equation (5), that is \( \rho_{44,41}(r, t) = \langle \hat{S}_{4d}(r, t)\hat{S}_{11}(0, t) \rangle \), a correlation between probability
Figure 3. The local (blue dashed line) and nonlocal (red solid line) FWM susceptibilities under different pumping detuning $\Delta_p/\gamma_{31}$. $\Gamma_{42} = \Gamma_{43} = 2\pi \times 3 \text{kHz}$, $\Gamma_{21} = \Gamma_{31} = 2\pi \times 6 \text{MHz}$, $\mu_{42} = \mu_{31} = 0.10 \text{eV}$, and $\mu_{43} = \mu_{32} = 0.23 \text{eV}$. $C_{0} = -2\pi \times 140 \text{GHz} \cdot \mu_{0}$, $\Delta_{c} = 50\gamma_{31}$, $\Delta_{d} = -50\gamma_{31}$, $\Omega_{c} = \Omega_{d} = 20\gamma_{31}$, and $\Omega_{p} = 0.1\gamma_{31}$. The atomic density $N_0 = 9.0 \times 10^{10} \text{cm}^{-3}$.

Figure 4. The atom-light part (blue dashed line) and the Rydberg–Rydberg part (red solid line) of the nonlinear coefficient under different atomic density $N_0$. $\Delta_{c} = 50\gamma_{31}$, $\Delta_{d} = -50\gamma_{31}$, and $\Delta_{p} = -50\gamma_{31}$. Other parameters are the same as in figure 3.

of the atom at $r$ staying on Rydberg state $|4\rangle$ and that of another Rydberg atom (at origin) jumping to the ground state. Since the two operators commute with each other, the same argument can be made when interchanging the two atoms. Of course, such transition from $|4\rangle$ to $|1\rangle$ is unpractical due to the same parity of two levels. However with the help of driven field ($\Omega_{d}$ in equation (5)), it manifests as a dominant part of the effective polarization and strengthens the FWM process.

Clearly, higher atomic density leads to shorter distance between atoms, thus significantly increases the two-body correlation and results in a much larger nonlinearity. In figure 4 we show the real part (a) and the imaginary part (b) of $\chi^{(3)}_R$ as a function of atomic density. Comparing with the local part of nonlinear coefficient, $\chi^{(3)}_a$ is proportional to $N_0$, while $\chi^{(3)}_R$ is proportional to $N_0^2$, see equations (6) and (12). One can verify the mechanism of the generation of $\Omega_s$ by examining the relation between the count rate of $\Omega_s$ (proportional to squared modulus of the nonlinear susceptibility [50, 55]) and the atomic density. The nonlinearity based on Rydberg–Rydberg interaction would manifest itself as a dependence of the count rate on $N_0^4$.

4. Conclusions

The FWM nonlinearity plays important roles in lots of applications in quantum information. And in this paper, we have investigated the enhancement of the third-order FWM nonlinearity via the correlation between two Rydberg atoms. Using the one-body and two-body elements of the density matrix, we solved the associated equations using perturbation method and show that the polarization which is induced by the pumping, coupling and driving field, and responsible for generating the signal field is composed of a local part from the interaction between atoms and photons and a nonlocal part due to the Rydberg–Rydberg
interaction. Using a Rydberg state with \( n = 60 \) as an example, we show via the numerical results that the nonlocal nonlinear susceptibility dominates in the total nonlinear coefficient. For the Rydberg state with a even larger quantum number, beside the above mentioned result, one finds that the Rydberg–Rydberg interaction becomes stronger and that leads to a larger blockade sphere. If we only focus on the two-body correlation, then a relatively low atomic density should be adopt and calculations show that the nonlinearity is actually weaker owning to the relation \( \chi^{(3)}_R \propto N_0^4 \). With a higher atomic density, the three-body correlation should be considered and the nonlinearity could be further enhanced.

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**Data availability statement**

No new data were created or analysed in this study.

**Appendix A. The equations for the one-body and two-body density-matrix elements**

For simplicity, we use complex decay rates \( g_{mn} \) in the following equations, and they are defined as the composed of decoherence rates and detunings. \( g_{21} = \gamma_{21} + i\Delta_p, g_{31} = \gamma_{31} + i\Delta_s, g_{41} = \gamma_{41} + i(\Delta_c + i\Delta_p), g_{32} = \gamma_{32} + i(\Delta_s - i\Delta_p), g_{42} = \gamma_{42} + i\Delta_c, g_{43} = \gamma_{43} + i\Delta_d \). The equations for one-body equations for

\[
\partial_t \rho_{11} = \Gamma_{21}\rho_{21} + \Gamma_{31}\rho_{33} - (i\rho_{12}\Omega_p + i\rho_{13}\Omega_s + c.c); \quad (A1)
\]

\[
\partial_t \rho_{22} = \Gamma_{42}\rho_{44} - \Gamma_{21}\rho_{22} + (i\rho_{23}\Omega_s - i\rho_{24}\Omega_c + c.c); \quad (A2)
\]

\[
\partial_t \rho_{33} = \Gamma_{43}\rho_{44} - \Gamma_{31}\rho_{33} - (i\rho_{34}\Omega_d + c.c); \quad (A3)
\]

\[
\partial_t \rho_{21} = -g_{21}\rho_{21} + i\Omega_p(\rho_{11} - \rho_{22}) - i\rho_{23}\Omega_s - i\rho_{24}\Omega_c^*; \quad (A4)
\]

\[
\partial_t \rho_{32} = -g_{32}\rho_{32} + i\rho_{31}\Omega_p^* - i\rho_{34}\Omega_c + i\rho_{35}\Omega_d^*; \quad (A5)
\]

\[
\partial_t \rho_{42} = -g_{42}\rho_{42} + i\Omega_c(\rho_{22} - \rho_{44}) + i\rho_{43}\Omega_d - i\rho_{44}\Omega_p^* - iN_a \int d^3r V(r)\rho_{44,42}(r, t); \quad (A6)
\]

\[
\partial_t \rho_{43} = -g_{43}\rho_{43} + i\rho_{41}\Omega_c + i\rho_{42}\rho_{44} - i\rho_{44}\Omega_c^* - iN_a \int d^3r V(r)\rho_{44,43}(r, t). \quad (A7)
\]

Equations for \( \rho_{31}, \rho_{42} \) and \( \rho_{41} \) are presented in the main text as equations (2a) and (2b), respectively. In the above equations, the phase-matching condition \( \Delta_s = \Delta_c + \Delta_p - \Delta_d \) is already assumed. Under the double perturbation we find that the \( \langle 1 \rangle \)-order equations for one-body elements are

\[
\begin{pmatrix}
-g_{21} & 0 & i\Omega_p^* \\
0 & -g_{31} & i\Omega_d^* \\
i\Omega_c & i\Omega_d & -g_{41}
\end{pmatrix}
\begin{pmatrix}
\rho_{11}^{(1)} \\
\rho_{21}^{(1)} \\
\rho_{31}^{(1)} \\
\rho_{41}^{(1)}
\end{pmatrix}
= 
\begin{pmatrix}
-i\Omega_p \\
0 \\
i\Delta_{44,41}
\end{pmatrix}. \quad (A8)
\]}
Here $\Delta_{44_{ij}} = N_{\alpha} \int d^3 r' V(r' - r) \rho_{44_{ij}}(r', r, t)$. In order to solve the (3)-order equations for the two-body elements we also need the value of (2)-order one-body element, and they reads

$$
\begin{pmatrix}
-\Gamma_{21} & 0 & -i\Omega_c & 0 & 0 & i\Omega_{c}^* & 0 & \Gamma_{42} \\
0 & -g_{23} & -i\Omega_d & 0 & 0 & 0 & i\Omega_{c}^* & 0 \\
-i\Omega_c^* & -i\Omega_d^* & -g_{24} & 0 & 0 & 0 & 0 & i\Omega_{c}^* \\
0 & 0 & 0 & -g_{32} & -i\Omega_c & i\Omega_{d}^* & 0 & 0 \\
0 & 0 & 0 & -\Gamma_{33} & -i\Omega_d & 0 & i\Omega_{d}^* & \Gamma_{43} \\
i\Omega_c & 0 & i\Omega_d & 0 & 0 & -g_{42} & 0 & -i\Omega_c \\
0 & i\Omega_c & 0 & i\Omega_d & 0 & 0 & -g_{43} & -i\Omega_d \\
0 & 0 & i\Omega_c & 0 & 0 & i\Omega_d & -i\Omega_{c}^* & -i\Omega_{d}^* & -\Gamma_{42} & -\Gamma_{43}
\end{pmatrix}
= \begin{pmatrix}
\rho_{21_{12}}^{(2)} \\
\rho_{31_{12}}^{(2)} \\
\rho_{41_{12}}^{(2)} \\
\rho_{21_{13}}^{(2)} \\
\rho_{31_{13}}^{(2)} \\
\rho_{41_{13}}^{(2)} \\
\rho_{21_{14}}^{(2)} \\
\rho_{31_{14}}^{(2)} \\
\rho_{41_{14}}^{(2)}
\end{pmatrix}
= \begin{pmatrix}
2\text{ Im}[\rho_{44_{12}}^{(1)}]\Omega_p \\
\rho_{44_{13}}^{(1)}\Omega_p \\
\rho_{44_{14}}^{(1)}\Omega_p
\end{pmatrix}
\tag{A9}
$$

The nonzero two-body elements on (2) order are the solutions to two set of closed equations. In matrix form, they are

$$
\begin{pmatrix}
M_1 & 0 & i\Omega_c^* & 0 & 0 & 0 & -i\Omega_c & 0 \\
0 & M_2 & i\Omega_d^* & 0 & 0 & 0 & 0 & -i\Omega_c \\
i\Omega_c & i\Omega_d & M_3 & 0 & 0 & 0 & 0 & -i\Omega_c \\
0 & 0 & 0 & M_4 & 0 & i\Omega_c^* & -i\Omega_c & 0 \\
0 & 0 & 0 & 0 & M_5 & i\Omega_c^* & 0 & 0 \\
0 & 0 & 0 & i\Omega_c & i\Omega_d & M_6 & 0 & 0 & -i\Omega_d \\
-i\Omega_c^* & 0 & 0 & -i\Omega_d^* & 0 & 0 & M_7 & 0 \\
0 & -i\Omega_c^* & 0 & 0 & -i\Omega_d^* & 0 & 0 & M_8 & i\Omega_d^* \\
0 & 0 & -i\Omega_c^* & 0 & 0 & -i\Omega_d^* & i\Omega_c & i\Omega_d & M_9
\end{pmatrix}
= \begin{pmatrix}
\rho_{21_{12}}^{(2)} \\
\rho_{31_{12}}^{(2)} \\
\rho_{41_{12}}^{(2)} \\
\rho_{21_{13}}^{(2)} \\
\rho_{31_{13}}^{(2)} \\
\rho_{41_{13}}^{(2)} \\
\rho_{21_{14}}^{(2)} \\
\rho_{31_{14}}^{(2)} \\
\rho_{41_{14}}^{(2)}
\end{pmatrix}
= \begin{pmatrix}
\rho_{44_{12}}^{(1)}\Omega_p \\
\rho_{44_{13}}^{(1)}\Omega_p \\
\rho_{44_{14}}^{(1)}\Omega_p
\end{pmatrix}
\tag{A10}
$$

With $M_1 = -g_{12} - g_{21}, M_2 = -g_{12} - g_{53}, M_3 = -g_{12} - g_{41}, M_4 = -g_{12} - g_{21}, M_5 = -g_{13} - g_{31}, M_6 = -g_{13} - g_{41}, M_7 = -g_{14} - g_{21}, M_8 = -g_{14} - g_{31}, M_9 = -g_{14} - g_{41}, M_{10} = -2g_{23}, M_{11} = -g_{21} - g_{31}, M_{12} = -g_{21} - g_{41}, M_{13} = -2g_{31}, M_{14} = -g_{31} - g_{41}, M_{15} = -2g_{41} - iV$.

The (3)-order equations depends on the above (2)-order elements. To obtain $\rho_{44_{11}}^{(3)}$ we need to solution a set of equations with 27 unknowns, and they are:

$$
\begin{pmatrix}
\rho_{22_{21}}^{(3)} \\
\rho_{23_{21}}^{(3)} \\
\rho_{24_{21}}^{(3)} \\
\rho_{32_{21}}^{(3)} \\
\rho_{33_{21}}^{(3)} \\
\rho_{34_{21}}^{(3)} \\
\rho_{42_{21}}^{(3)} \\
\rho_{43_{21}}^{(3)} \\
\rho_{44_{21}}^{(3)}
\end{pmatrix}
= \begin{pmatrix}
\rho_{21_{12}}^{(2)} \\
\rho_{31_{12}}^{(2)} \\
\rho_{41_{12}}^{(2)} \\
\rho_{21_{13}}^{(2)} \\
\rho_{31_{13}}^{(2)} \\
\rho_{41_{13}}^{(2)} \\
\rho_{21_{14}}^{(2)} \\
\rho_{31_{14}}^{(2)} \\
\rho_{41_{14}}^{(2)}
\end{pmatrix}
= \begin{pmatrix}
-\rho_{44_{12}}^{(1)}\Omega_p \\
\rho_{44_{13}}^{(1)}\Omega_p \\
\rho_{44_{14}}^{(1)}\Omega_p
\end{pmatrix}
\tag{A11}
$$

They are obtained following the same procedure of finding the matrices of (2)-order equations. The matrix equation are not shown here since the size of the matrix is too large.

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