Phase Modulation in Rydberg Dressed Multi-Wave Mixing processes

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We study the enhancement and suppression of different multi-waving mixing (MWM) processes in a Rydberg-EIT rubidium vapor system both theoretically and experimentally. The nonlinear dispersion property of hot rubidium atoms is modulated by the Rydberg-Rydberg interaction, which can result in a nonlinear phase shift of the relative phase between dark and bright states. Such Rydberg-induced nonlinear phase shift can be quantitatively estimated by the lineshape asymmetry in the enhanced and suppressed MWM processes, which can also demonstrate the cooperative atom-light interaction caused by Rydberg blockaded regime. Current study on phase shift is applicable to phase-sensitive detection and the study of strong Rydberg-Rydberg interaction.

The phase modulation as well as the refractive index modification in a Rydberg medium, caused by electric fields produced either externally or internally owing to the interparticle interactions, is of central importance in nonlinear optics, laser technology, quantum optics and optical communications1. Because the high-lying Rydberg electron is very far from the core of the atom, the atom possesses exaggerated properties, such as huge polarizability that scales as \( n^7 \), where \( n \) is the principal quantum number. These properties lead to strong and tunable Rydberg-Rydberg interactions2–4 among the atoms, which can render the Rydberg medium intrinsically nonlinear. For example, Rydberg electromagnetically induced transparency (EIT) makes the transmission through the medium highly sensitive to electric fields1, which can enable modifications on the refractive index and nonlinear phase shift due to the interparticle interactions in the nonlinear processes associated with EIT.

Comparing with the other nonlinear optical processes, the multi-waving mixing (MWM) processes in Rydberg-EIT medium have unique features5–8, and one typical feature is that the coherence time of the generated signal is shorter than the time of ionization9, while it is known that the incoherence plasma formation in Rydberg gases is \( \sim 100 \text{ ns} \) or longer10,11. With EIT configuration, the coherence between the ground state and highly-excited Rydberg states is well established, which can enhance the efficiency of the MWM processes12,13. In addition, the spatial arrangement of EIT configuration will suppress the Doppler width greatly, which makes atoms in the beam volume behave like cold atoms with reduced Doppler effect14–17. Finally, the EIT windows will pick up the corresponding MWM signals with narrow linewidth (less than 30 MHz). Therefore, probing the EIT-assisted MWM processes can provide a powerful spectral method to investigate the properties of Rydberg atoms.

In this paper, we study the enhancement and suppression of Rydberg dressed MWM processes with the assistance of EIT windows in a hot Rb atomic system both theoretically and experimentally. The enhanced and suppressed MWM signals are significantly modified via the relative phase control18 due to the nonlinear dispersion property modification induced by corresponding dressing effects and the cooperative nonlinear effect19,20 from the Rydberg blockaded regime. The introducing of strong Rydberg-Rydberg
Figure 1. (a) A five-level atomic system in Rydberg-EIT rubidium atom for dressed MWM processes. (b) Experimental setup for different MWM processes. L-lens, D-detector, FD-frequency doubler, HW-half wave plate with corresponding wavelength, PBS-polarized beam splitter with corresponding wavelength. Double-headed arrows and filled dots denote horizontal polarization and vertical polarization of the incident beams, respectively. (c) Theoretical calculations corresponding to the change in refractive index ($\Delta n_{\text{r}}$) of a medium for a probe laser (or MWM signal) frequency versus $\Delta_{1}$ and $\Delta_{2}$: $\Omega_{1} = 2\pi \times 54$ MHz, $\Omega_{2} = 2\pi \times 7.6$ MHz, $\Omega_{1} = 2\pi \times 142$ MHz, $\Omega_{4} = 2\pi \times 224$ MHz. The atom density is $1.0 \times 10^{13}$ cm$^{-3}$.

interactions into atom-light interaction means that each atom can no longer be treated independently and the correlations between atoms must be taken into consideration, which can be interpreted as a cooperative effect. The cooperative nonlinearity in an atomic ensemble can be much more obvious for high atomic density. As a result, the spatial effects of corresponding dressed signals can visually advocate the change of nonlinear dispersion property in current experiment. The intensity evolutions of enhancement and suppression results may map onto the nonlinear phase shift in modulated dispersion property by scanning dressing fields. Different from the asymmetry degree of cavity transmission profile method, such nonlinear phase shift with background-free advantages can be estimated via the dressing asymmetry in enhanced and suppressed MWM linshapes, which can also demonstrate the excitation blockade effects. The nonlinear phase shift of the relative phase between dark and bright states gives a novel way for studying the Rydberg-Rydberg interactions and phase-sensitive detection.

Results
An X-type five-level $^{85}$Rb atomic system, consisting of two hyperfine states F = 3 ($|0\rangle$) and F = 2 ($|3\rangle$) of the ground state 5S$_{1/2}$, a first excited state 5P$_{3/2}$ ($|1\rangle$), a lower-lying excited state 5D$_{3/2}$ ($|2\rangle$), and a highly-excited Rydberg state nD$_{3/2}$ ($|4\rangle$), is used to generate the EIT-assisted MWM processes. Six laser beams derived from four commercial external cavity diode laser systems with frequency-stabilized servos are coupled into the corresponding transitions as shown in Fig. 1(a). The experimental setup is shown in Fig. 1(b). Except for the $E_{4}$', the experimental setup is essentially the same as previous work. A weak laser beam $E_{1}$ (780.24 nm with a diameter of 0.8 mm, frequency $\omega_{1}$, wavevector $k_{1}$) from LD1 probes the lower transition $|0\rangle$ to $|1\rangle$, while a pair of coupling beams $E_{3}$ (780.23 nm, $\omega_{3}$, $k_{3}$) and $E_{4}$' ($\omega_{4}$, $k_{4}'$), derived from the same LD3 with a small angle between them both with the same diameter of 1 mm, connect another lower transition $|3\rangle$ to $|1\rangle$. To excite hot rubidium atoms from level $|1\rangle$ to Rydberg states $|2\rangle$, we obtain the needed 480 nm laser $E_{2}$ ($\omega_{2}$, $k_{2}$) by the way of frequency doubling LD2 at $\approx 960$ nm. The strong beam $E_{2}$ (diameter 1 mm) adding onto the beam $E_{1}$ (in the same direction), which counter-propagates with beam $E_{1}$, drives the highly-excited Rydberg transition $|1\rangle$ to $|2\rangle$. $E_{4}$ (775.98 nm with a diameter of 1 mm, frequency $\omega_{4}$, wavevector $k_{4}$) and $E_{4}'$ ($\omega_{4}'$, $k_{4}'$) from LD4 drive the transition $|1\rangle$ to $|4\rangle$.

Different-order dressed MWM processes can be obtained by turning the incident beams on selectively. First, by blocking beams $E_{1}$ and $E_{4}$', a four-wave mixing (FWM) process $E_{\text{FWM1}}$ with the phase-matching condition (PMC) $k_{\text{FWM1}} = k_{1} + k_{4} - k_{4}'$ can be dressed by $E_{2}$ in the Y-type four-level subsystem $|0\rangle$ $\rightarrow$ $|1\rangle$ $\rightarrow$ $|2\rangle$ $\rightarrow$ $|4\rangle$. Next, when opening all other beams except $E_{4}$', a non-EIT-assisted FWM process $E_{\text{FWM2}}$ with $k_{\text{FWM2}} = k_{1} + k_{4} - k_{4}'$ in the $\Lambda$-type three-level subsystem $|0\rangle$ $\rightarrow$ $|1\rangle$ $\rightarrow$ $|3\rangle$ and two EIT-assisted six-wave mixing (SWM) processes $E_{\text{SWM1}}$ involving in Rydberg states and $E_{\text{SWM2}}$ (with the PMCs of $k_{\text{SWM1}} = k_{1} + k_{3} - k_{4}$ and $k_{\text{SWM2}} = k_{1} + k_{3} - k_{4}' + k_{4} - k_{3}$) can be observed in $|0\rangle$ $\rightarrow$ $|1\rangle$ $\rightarrow$ $|3\rangle$ $\rightarrow$ $|2\rangle$ and $|0\rangle$ $\rightarrow$ $|1\rangle$ $\rightarrow$ $|3\rangle$ $\rightarrow$ $|4\rangle$, respectively. These MWM signals have the same emitting direction (opposite to the direction of $E_{4}'$, as shown in Fig. 1(b)) except for $E_{\text{FWM1}}$ (propagating along the opposite direction of $E_{4}'$). The various MWM processes are identified by tuning the frequency detuning of corresponding coupling beams and detected by respective avalanche photodiode detectors (APD). Specifically, the MWM processes related to the Rydberg state $|2\rangle$ may be called Rydberg MWM signals with strong Rydberg-Rydberg interactions.

\begin{align*}
\text{Figure 1. (a) A five-level atomic system in Rydberg-EIT rubidium atom for dressed MWM processes. (b) Experimental setup for different MWM processes. L-lens, D-detector, FD-frequency doubler, HW-half wave plate with corresponding wavelength, PBS-polarized beam splitter with corresponding wavelength. Double-headed arrows and filled dots denote horizontal polarization and vertical polarization of the incident beams, respectively. (c) Theoretical calculations corresponding to the change in refractive index ($\Delta n_{\text{r}}$) of a medium for a probe laser (or MWM signal) frequency versus $\Delta_{1}$ and $\Delta_{2}$: $\Omega_{1} = 2\pi \times 54$ MHz, $\Omega_{2} = 2\pi \times 7.6$ MHz, $\Omega_{1} = 2\pi \times 142$ MHz, $\Omega_{4} = 2\pi \times 224$ MHz. The atom density is $1.0 \times 10^{13}$ cm$^{-3}$.}
\end{align*}
The interaction among Rydberg atoms scales with $n^{1/2}$ and leads to the change in refractive index of the medium and nonlinear phase shift of the relative phase between dark and bright states, which can be mapped onto the enhancement and suppression of EIT-assisted MWM processes with dressing effects. To be specific, the modification of refractive index ($n_r$) caused by Rydberg energy level shift ($\Delta \omega_2$) can be expressed as

$$\Delta n_r = \left( \frac{\partial n_r}{\partial \omega_2} \right) \Delta \omega_2,$$

where $\frac{\partial n_r}{\partial \omega_2} = (n_x - 1)/\omega_2$, is the Rydberg state coupling laser frequency and $n_x$ is the group refractive index. The theoretical simulation of $\Delta n_r$ is shown in Fig. 1(c). The phase modulation ($\Delta \Phi_t$) due to the strong cooperative atom-light interaction due to Rydberg blockade is described as

$$\Delta \Phi_t(U) = \left( L \omega_2 \Delta n_r / c \right),$$

which means the phase shift is proportional to the Rydberg induced dispersion change $\Delta n_r$ and the propagation distance $L$ (or equivalently atomic density). See Methods for the theoretical derivations of $\Delta n_r$ and $\Delta \Phi_t(U)$.

For the two FWM signals (via the pathways $\rho_{50}^{(0)} \rightarrow \rho_{10}^{(0)} \rightarrow \rho_{50}^{(2)} \rightarrow \rho_{10}^{(2)} \rightarrow \rho_{10}^{(FWM1)}$ and $\rho_{50}^{(0)} \rightarrow \rho_{10}^{(0)} \rightarrow \rho_{50}^{(3)} \rightarrow \rho_{10}^{(FWM2)}$) with the dressing effects of $E_2$ and $E_6$, the corresponding third-order polarizations $P^{(3)}$ for the output FWM signals under steady-state condition are given by

$$P^{(3)}_{FWM1} = \int_{-\infty}^{+\infty} \frac{i N_0 d_{i0} \Omega_1 \left| \Omega_2 \right|^2 N(\nu) d\nu}{\gamma_1 + \left( \left| \Omega_2 \right| / n \right)^{1/4} e^{i \Delta \Phi / \gamma_2} + \left| \Omega_4 \right|^{1/2} e^{i \Delta \Phi' / \gamma_4}},$$

$$P^{(3)}_{FWM2} = \int_{-\infty}^{+\infty} \frac{i N_0 d_{i0} \Omega_1 \left| \Omega_3 \right|^2 N(\nu) d\nu}{\gamma_1 + \left( \left| \Omega_2 \right| / n \right)^{1/4} e^{i \Delta \Phi / \gamma_2} + \left| \Omega_4 \right|^{1/2} e^{i \Delta \Phi' / \gamma_4}},$$

where $N(\nu) = N_0 \exp(-\nu^2/\sigma^2)/\sqrt{\pi}^{1/2}$ is the particle number density in terms of speed distribution function; $\gamma_1 = d_i E_1/\hbar$ ($i = 1, 2 \ldots$) is the Rabi frequency between $|i\rangle \leftrightarrow |j\rangle$; $d_i$ is the dipole moment; $N_0$ is the atom density; $\gamma_2 = (\Gamma_{20} + \Gamma_\nu + i(\Delta_1 + k_1 \nu)$, $\gamma_3 = (\Gamma_{30} + \Gamma_\nu + i(\Delta_2 + k_2 \nu)$. $\gamma_4 = (\gamma_1 + \gamma_2) / n$ and $\gamma_5 = (\gamma_1 + \gamma_2 + \gamma_4 / n) / n$. The corresponding transition frequency $\omega_1$ and the laser frequency $\omega_0$ of $E$. Note that the collision ionization rate $\Gamma_{i\nu}$, transit time $\Gamma_\nu$ and the Doppler effect ($k \nu$) should be considered. For the EIT-assisted SWM signals via $\rho_{50}^{(0)} \rightarrow \rho_{10}^{(0)} \rightarrow \rho_{50}^{(2)} \rightarrow \rho_{10}^{(2)} \rightarrow \rho_{10}^{(FWM1)}$ and $\rho_{50}^{(0)} \rightarrow \rho_{10}^{(0)} \rightarrow \rho_{50}^{(3)} \rightarrow \rho_{10}^{(FWM2)}$, the corresponding fifth-order polarizations $P^{(5)}$ are given by

$$P^{(5)}_{SWM1} = \int_{-\infty}^{+\infty} \frac{i N_0 d_{i0} \Omega_1 \left| \Omega_2 \right|^2 \left| \Omega_4 \right|^2}{\gamma_1 + \left( \left| \Omega_2 \right| / n \right)^{1/4} e^{i \Delta \Phi / \gamma_2} + \left| \Omega_4 \right|^{1/2} e^{i \Delta \Phi' / \gamma_4}},$$

$$P^{(5)}_{SWM2} = \int_{-\infty}^{+\infty} \frac{i N_0 d_{i0} \Omega_1 \left| \Omega_3 \right|^2 \left| \Omega_4 \right|^2}{\gamma_1 + \left( \left| \Omega_2 \right| / n \right)^{1/4} e^{i \Delta \Phi / \gamma_2} + \left| \Omega_4 \right|^{1/2} e^{i \Delta \Phi' / \gamma_4}},$$

Here, the additional phase factors $e^{i \Delta \Phi}$ and $e^{i \Delta \Phi'}$ are introduced into the dressing terms $\left| \Omega_2 \right| / n^{1/4}$ and $\left| \Omega_4 \right|^{1/2}$ to account for the propagation effect. $\Delta \Phi = \Delta \Phi_t + \Delta \Phi_d$, where $\Delta \Phi_t(U)$ is the phase modulation induced by the possibly coherent Rydberg-Rydberg interaction $U$; the relative phase $\Delta \Phi_t$ and $\Delta \Phi'$ are related to the orientations of induced dipole moments and can be manipulated by corresponding laser frequency detuning and Rabi frequency.
Phase modulated intensity and spatial effects in the Y-type subsystem. Figure 2 shows the dressed FWM1 process in the Y-type four-level subsystem $\ket{0}\leftrightarrow\ket{1}\leftrightarrow\ket{2}\leftrightarrow\ket{4}$ by scanning the frequency of Rydberg coupling field $E_2$. Suppressed and enhanced FWM1 signals (the suppressed condition is $\Delta_1 + \Delta_4 = 0$ and the enhanced condition is $\Delta_1 + \Delta_4 = (\Delta_1/2 + \sqrt{\Delta_1^2 + 4|\Omega_4|^2}/2) = 0$) are observed by changing the frequency detuning of $E_1$ or $E_2$. According to the new two-photon dressed rule, the moving states $\ket{\pm}$ will impose influence on the enhancing and suppressing results of MWM signals. Let’s first show the generating process of Rydberg-dressing enhancement and suppression simply. Figure 2(a) shows the switch from an enhanced peak to a suppressed dip by growing $\Delta_1$ at $\Delta_1 = -\Delta_3 = 30$ MHz. The dressing processes can be considered as following: first, as shown in Fig. 2(f), level $\ket{1}$; and then $\ket{1}$ is split into $\ket{1\pm\gamma}$ secondly by $E_2$. Therefore, once the dressing level $\ket{1\pm\gamma}$ moved around the position of $\Delta_i$, the suppressed condition is satisfied and the suppressed case of FWM1 occurs in Fig. 2(a). Figure 2(b) shows the dependence of suppressed dip on the strength of $E_i$ at $\Delta_1 = \Delta_4 = 0$. The enhanced condition cannot be satisfied in the situation shown in Fig. 2(b) in which the suppressed dip increases as the power of $E_i$ increases and the two-step dressing process can be simplified as level $\ket{1}$ is split by $E_i$. In order to visually investigate the nonlinear dispersion property induced by Rydberg dressing effect and cooperative effect, we turn to the spatial effects of the images of dressed signals. With only $E_1$ and $E_2$ turned on, Fig. 2(c) shows the focusing/defocusing effects of probe signal versus $\Delta_1$. Nonlinear refractive index $n_r$ is negative in the self-focusing medium ($\Delta_1 < 0$) while positive in the self-defocusing one ($\Delta_1 > 0$). Figure 2(d1,d2) show the probe images with $E_1\&E_2$ and $E_1\&E_2\&E_4$ on versus $\Delta_1$, respectively. With $E_2$ blocked, the focusing/defocusing effects of probe images at different $\Delta_1 + \Delta_4 = 0$ can be stronger than the effects in Fig. 2(c) due to the growing of absolute value of refractive index. With $E_1\&E_2\&E_4$ on, the images of dressed $E_2$ EIT become more defocusing compared with the corresponding ones in Fig. 2(d1) due to $\Delta n_{\pm}$ is negative in most part of the resonance line as shown in Fig. 1(c). In addition, the spatial splitting and shift in Fig. 2(d2) can be attributed to $\delta k_\perp = \partial (\Delta \Phi) / \partial \xi$, where $\Delta \Phi$ can be modified as

\begin{align}
\delta k_\perp &= \partial (\Delta \Phi) / \partial \xi \\
\delta k_\perp &= \Delta \Phi / \xi \\
\delta k_\perp &= \Delta \Phi / \xi \\
\delta k_\perp &= \Delta \Phi / \xi \\
\end{align}
\[ \Delta \Phi_1(U) = L \omega \Delta n e^{-\Delta^2}/c. \]  

Equation (4) by setting \( \Delta \) factor as of dressed MWM signals. To estimate such dressing asymmetry quantitatively, we define the asymmetry on FWM2 versus \( E \), setting \( \Delta \) at discrete with the SWM2 by scanning shift induced by the Rydberg blockade. Figure 3 shows the enhanced and suppressed FWM2 coexisting to the one-photon peak of the FWM1 signal versus \( \gamma \) and can be described by the single-photon term \( \gamma_1 \) in Eq. (3). The intensity of FWM1 in Fig. 2(g) is first suppressed and then enhanced at \( \Delta_v = 32 \text{MHz} \), while it is first enhanced and then suppressed at \( \Delta_v = 32 \text{MHz} \). Obviously, a dressing asymmetry occurs with \( \Delta_v = 0 \) considered as a center.

In general, the dressing enhancement peaks and suppression dips are symmetrical distributed along the center. However, the induced nonlinear phase shift may lead to the asymmetry in the lineshapes of dressed MWM signals. To estimate such dressing asymmetry quantitatively, we define the asymmetry factor as

\[ A_F = (e_2 - e_1 + s_2 - s_1)/(e_1 + e_2 + s_1 + s_2), \]

where \( e_i \) and \( s_i \) represent the enhancement and suppression of FWM1 intensity; subscripts 2 and 1 indicate \( e_i \) (or \( s_i \)) are taken with \( \Delta_v > 0 \) and \( \Delta_v < 0 \), respectively. Actually, the relationship between \( A_F \) and phase shift can be described as

\[ A_F \propto (\Delta_{\text{FWHM}}/\beta) (\alpha_1 \Delta \Phi + \alpha_2 \Delta \Phi'), \]

where \( \Delta_{\text{FWHM}} \) and \( \beta \) are the full width at half maximum (FWHM) and full width at a certain frequency detuning point of the corresponding profile, respectively; \( \alpha_1 \) and \( \alpha_2 \) are the ratio parameters for phase shift \( \Delta \Phi \) and \( \Delta \Phi' \) caused by \( E_1 \) and \( E_p \) respectively.

According to Eq. (8), the value of \( A_F \) in Fig. 2(g) is about 0.58 at \( |\Delta_v| = 32 \text{MHz} \). Due to the absence of Autler-Townes (AT) splitting on the profile, the dressing effect of \( E_2 \) on the one-photon term \( \gamma_1 \) that only affects the intensities of the signals can be neglected. Since the modulated results of FWM1 in Fig. 2(g) are related to the change in \( \Delta_v \), one can attribute the results to the dressing effect of \( E_2 \) on \( \gamma_1 \). Therefore, \( A_F \) in Fig. 2(g) is mainly contributed by the Rydberg dressing and cooperative nonlinear effect. The denominator of Eq. (3) is simplified to \( \gamma_1 + (|\Omega_2|/|\Omega_1|) \) \( \phi_4 \gamma_2 \) and can explain Fig. 2(g) well by setting \( \Delta \Phi = \Delta \Phi_1(U) + \Delta \Phi_2 = -\pi/3 \). (see Fig. 2(g)).

Figure 2(h) is the modulated enhancement and suppression of FWM1 signal by increasing \( \Delta_v \) at \( \Delta_v = 0 \), and \( A_F \) is about 0.91 at \( |\Delta_v| = 50 \text{MHz} \). Different from the case in Fig. 2(a), the Lorentzian profile (curve constituted of the baseline of each signal) is a two-photon peak of the FWM1 signal versus \( \Delta_v \), which can be described by the two-photon term \( \gamma_1 \) in Eq. (3). Obviously, the change of \( \Delta_v \) can also affect the modulated results of FWM1, and it can be ascribed to the dressing effect of \( E_2 \) on \( \gamma_1 \) associating with self-dressing shown in Eq. (3). As a consequence, the denominator of Eq. (3) is simplified as

\[ \gamma_1^2 + (|\Omega_2|/|\Omega_1| \phi_4 \gamma_2) \] \( \gamma_1 + (|\Omega_2|/|\Omega_1|) \phi_2 \) \( \gamma_2 \) \} \] which can account for Fig. 2(h) with \( \Delta \Phi = \Delta \Phi_1(U) + \Delta \Phi_2 = -\pi/3 \) and \( \Delta \Phi' = -\pi \). (see Fig. 2(h)).

**Phase modulated intensity in the inverted-Y type subsystem.** Now, we try to pick out the phase shift induced by the Rydberg blockade. Figure 3 shows the enhanced and suppressed FWM2 coexisting with the SWM2 by scanning \( \Delta_v \) at discrete \( \Delta_v \). To be specific, Fig. 3(a) is the case with \( E_2 \) beam blocked and shows the dressing effect of \( E_1 \) on FWM2 versus \( \Delta_v \) at different \( \Delta_v \), which can be well simulated by Eq. (4) by setting \( \Delta \Phi' = -\pi/6 \) at \( \Delta_v = 150 \text{MHz} \) (see Fig. 3(a1)). As defined above, the dressing asymmetry factor \( A_F \) in Fig. 3(a) is 0.19 at \( |\Delta_v| = 80 \text{MHz} \). The profile (curve constituted of the baseline of each signal) in Fig. 3(a) is the one-photon peak of FWM2 signal versus \( \Delta_v \) (see the one-photon term \( \gamma_1 \) in Eq. (4)) with \( E_2 \) blocked, and the peak is broadened to 200 MHz by the Doppler effect \( \Delta_v = k_v v + k_v \). Figures 3(b,c) are the ones with the dressing effect of \( E_2 \) (coupling the transition between \( 5p_{32} \leftrightarrow 54D_{32} \)) at different atomic densities, respectively. The profiles in Figs. 3(b, c) increase to be as high as 0.61 and 0.86 at \( |\Delta_v| = 80 \text{MHz} \) due to the introducing of Rydberg field. The difference between the asymmetry factors on the profiles can be explained by the nonlinear phase shift caused by \( E_2 \) dressing effect and the cooperative atom-light interaction\(^{22}\). Since both Fig. 3(b,c) are related to the same Rydberg state \( 54D_{32} \), the phase shift induced by the change of cooperative nonlinearity due to Rydberg-Rydberg interaction can be observed by comparing the modulated results of \( N_{\text{r}} = 1 \times 10^{12} \text{cm}^{-3} \) and \( N_{\text{r}} = 2.4 \times 10^{12} \text{cm}^{-3} \). The introducing of correlations between atoms into atom-light interaction can
The phase shift difference in inverted-Y subsystem with optical pumping effect by changing the values of $\Delta_1$, which is broadened by the Doppler effect $\Delta_1 - \Delta_2 = k_{y1} + k_{y2}$. (a1)(b1) and (c1) are the theoretical predictions corresponding to (a)(b) and (c), respectively. (a1) $\Delta \Phi' = -\pi/6$. (b1) $\Delta \Phi' = -\pi/6$, $\Delta \Phi = \Delta \Phi_1 + \Delta \Phi_2 = -\pi/12$. (c1) $\Delta \Phi' = -\pi/6$, $\Delta \Phi = \Delta \Phi_1 + \Delta \Phi_2 = -\pi/3$, $\Delta_2 = 0$, $\Delta_3 = 150$ MHz. $\Omega_1 = 2\pi \times 54$ MHz at 0.5 mW, $\Omega_2 = 2\pi \times 7.6$ MHz at 200 mW, $\Omega_3 = 2\pi \times 116$ MHz at 4 mW, $\Omega_4 = 2\pi \times 170$ MHz at 5 mW, $\Omega_5' = 2\pi \times 275$ MHz at 13 mW.

Figure 3. The change in $\Delta_1$ induced enhancement and suppression of FWM2 together with the SWM2 process by scanning $\Delta_1$, (a) without $E_l$ and (b) with $E_l$ coupling the transition between 5P$_{3/2}$ and 5P$_{1/2}$, respectively, at atom density $N_0 = 1 \times 10^{12}$ cm$^{-3}$. (c) is the same to (b) except for $N_0 = 2.4 \times 10^{12}$ cm$^{-3}$. The profile (curve constituted of the baseline of each signal) in each panel is the FWM2 signal versus $\Delta_1$, which is broadened by the Doppler effect $\Delta_1 - \Delta_2 = k_{y1} + k_{y2}$. (a1)(b1) and (c1) are the theoretical predictions corresponding to (a)(b) and (c), respectively. (a1) $\Delta \Phi' = -\pi/6$. (b1) $\Delta \Phi' = -\pi/6$, $\Delta \Phi = \Delta \Phi_1 + \Delta \Phi_2 = -\pi/12$. (c1) $\Delta \Phi' = -\pi/6$, $\Delta \Phi = \Delta \Phi_1 + \Delta \Phi_2 = -\pi/3$, $\Delta_2 = 0$, $\Delta_3 = 150$ MHz. $\Omega_1 = 2\pi \times 54$ MHz at 0.5 mW, $\Omega_2 = 2\pi \times 7.6$ MHz at 200 mW, $\Omega_3 = 2\pi \times 116$ MHz at 4 mW, $\Omega_4 = 2\pi \times 170$ MHz at 5 mW, $\Omega_5' = 2\pi \times 275$ MHz at 13 mW.

lead to a cooperative effect. The increase of Rydberg atom population will increase the cooperative nonlinearity and result in a dramatically change of the measured lineshapes.

Comparing the fourth curve in Fig. 3(b) with the fourth one at $\Delta_1 = 80$ MHz in Fig. 3(a), the difference between the modulated results can be explained well by setting $\Delta \Phi = \Delta \Phi_1 + \Delta \Phi_2 = -\pi/12$ (see Fig. 3(b1)). For the higher density shown in Fig. 3(c), the theoretical prediction agrees well with the experimental results by setting $\Delta \Phi = \Delta \Phi_1 + \Delta \Phi_2 = -\pi/3$ (see Fig. 3(c1)). Obviously, the phase shift as well as the dressing asymmetry factor grows with the atomic density and such density-dependent characteristic can demonstrate the $\Delta \Phi_1$ caused by the change of cooperative nonlinearity. Considering that the values of $\Delta \Phi_2$ in Fig. 3(b,c) are almost same due to the saturated dressing effect, the phase difference caused by the increase of cooperative nonlinear effect is approximately $\pi/4$. Therefore, such results sufficiently prove the existence of the phase shift induced by the interaction between Rydberg atoms.

Besides of the blockade dressed SWM process discussed above, one can further use the Rydberg MWM process to study the phase shift induced by the strong Rydberg-Rydberg interaction. Figures 4(a,b) show the induced enhancements and suppressions of FWM2 and SWM1 together with the SWM2 processes for 37D and 54D by varying $\Delta_1$ at $\Delta_2 = \Delta_3 = 0$, respectively. The peaks of the FWM2 and SWM1 signals versus $\Delta_1$ for 37D and 54D are shown by the Lorentzian profiles, which can be described by the one-photon term $\gamma_1$ in Eqs. (4) and (5), respectively. Since the results are related to the changing of $\Delta_1$, they can be attributed to the dressing effects on the one-photon term $\gamma_1$ as shown in Eqs. (4) and (5). The phase shift of $\Delta \Phi$ on the dressing term $|\Omega_1|^2/\gamma_1$ is $-\pi/6$ (see Fig. 4(a1,b1)). The difference of the phase shifts induced by different cooperative nonlinear effect for the two principal quantum numbers can be obtained by comparing the corresponding modulated results at the same frequency detuning. In the current case, a phase shift difference of $\pi/4$ is introduced between 37D and 54D due to the $n$-dependent characteristic of cooperative nonlinearity.

Figure 4(c,d) are the enhanced and suppressed SWM1 for 37D and 54D at $\Delta_1 = -\Delta_2 = 30$ by altering $\Delta_2$, respectively. The Lorentzian profiles with linewidth of 60 MHz are the two-photon peaks of the SWM1 signal versus $\Delta_2$ for 37D and 54D, respectively, and related to the two-photon term $\gamma_2$ in Eq. (5) (see Fig. 4(c1,d1)). Different from the former cases, we are now interested in the dressing effects on the two-photon term $\gamma_2$ whereas the dressing effects on $\gamma_1$ can be neglected. However, except for the increase of suppression in correspondingly modulated SWM1 signals of 37D and 54D, the dressed results are almost the same for both states due to the strong optical pumping. Therefore, the information of the phase difference in inverted-Y subsystem with optical pumping effect by changing $\Delta_2$ is not as...
obvious as in Y-type system by changing Δ4. Here, we have to mention that the central frequency shift of the Lorentzian profiles is observed due to the energy shift induced by different Rydberg-Rydberg interactions.

Finally, we characterize the blockaded enhancement and suppression results at Δ4 = −120 MHz in Fig. 4(a,b) as the functions of the probe field strength P4, the Rydberg state coupling field strength P3, and the coupling field strength P4 for three nD5/2 states. We expand Eqs. (3)–(6) as Taylor series based on the dressing fields. Taking Eq. (5) as an example, we have

\[ P_{0}^{(5)}_{\text{SWM1}} = \Omega_{\text{a}} \int_{-\infty}^{+\infty} \frac{\Omega_{a} N(v)dv}{1 + (\frac{\Omega_{a}}{n^{1.04}})^{2} e^{2i\Delta\Phi} / \gamma_{1}\gamma_{2} + \Omega_{a}^{2} e^{2i\Delta\Phi'} / \gamma_{1}\gamma_{4}^{3}} + \frac{+\infty}{-\infty} + 12(\frac{\Omega_{a}}{n^{1.04}})^{2} e^{i(\Delta\Phi+\Delta\Phi')} / \gamma_{1}\gamma_{2}\gamma_{4} + 3(\frac{\Omega_{a}}{n^{1.08}})^{2} / \gamma_{1}\gamma_{2} + 3\Omega_{a}^{4} / \gamma_{1}\gamma_{4}^{2} \right] 

where \( \Omega_{a} = i N_{0}^{0.2} d_{10}^{0.2} \Omega_{1}^{0.2} / \Omega_{2}^{0.4} / n^{1.04} \right|^{2} / \gamma_{1}^{3} \gamma_{2} \gamma_{3}^{3} \). Therefore, the intensities of the enhanced peak, suppressed dip and background are related to the terms \( \Omega_{a} \right|^{2} / \gamma_{1}^{3} \gamma_{2} \gamma_{3}^{3} \). Consequentially, the Rydberg dressed signals for each principal quantum number n are scaled to \( n = 37 \) by the factor \((n/37)^{2}\) accounting for the decrease in \( d_{10} \) with increasing n. Here, \( n = n - \delta \), and \( \delta = 1.35 \) is due to the quantum defect for nD5/2 state27.

Figure 5(a) presents the \( E_{1} \) power dependences of the (a1) enhanced peak, (a2) suppressed dip and (a3) background, respectively, for three nD5/2 states. The change of enhanced peak is mainly contributed by the enhanced FWM2 & SWM1 processes and the two-photon peak of SWM2. The trend of the suppressed dip can be understood as the saturating dressing-effect of \( E_{2} \) at \( \Delta_{1} + \Delta_{4} = 0 \). The background evolution is due to the sum of FWM2 and SWM1 processes. Based on the evolutions of enhancement and suppression, we can draw the conclusion that the dressing asymmetry \( A_{2} \) increases with the strength of \( E_{1} \). The saturating dressing-effect of \( E_{2} \) means the phase shift is mainly caused by the existence of \( E_{2} \) dressing and blockaded effect.

![Figure 4](https://example.com/image4.png)

Figure 4. Dressed MWM processes by scanning \( \Delta_{4} \). (a-b) The enhancement and suppression of FWM2 and SWM1 dressed by the SWM2 process with \( \Delta_{4} \) growing at \( \Delta_{1} = \Delta_{2} = 0 \) for 37D and 54D, respectively. The Lorentzian profiles (curve constituted of the baseline of each signal) are the FWM2 and SWM1 signal versus \( \Delta_{4} \) for 37D and 54D, respectively. (c-d) The enhanced and suppressed SWM1 by increasing the \( \Delta_{4} \) with \( \Delta_{1} = \Delta_{2} = 30 \) MHz for 37D and 54D, respectively. The Lorentzian profiles are the SWM1 signal versus \( \Delta_{4} \) for 37D and 54D. (a1-d1) are the theoretical curves corresponding to (a-d) with \( \Delta\Phi' = -\pi/6 \), respectively. The Rydberg-induced phase shift difference between 37D and 54D is about \( \pi/4 \).
The cases of the $E_2$ power dependences for three $nD_{5/2}$ states are shown in Fig. 5(b). First, we focus on the $P_2$ dependence of the enhanced peak (see Fig. 5(b1)). At the low excitation intensity, the enhanced FWM2 signal and SWM2 signal contribute to the enhanced peak. As $\Omega_2$ increases, the enhanced SWM1 signal also makes the height of enhanced peak increase. However, the blockade term ($|\Omega_2|^2/n_1^{11/2}$) makes the curve saturated at higher power level. Then, the descending part of the curve is due to the dressing effect of $E_2$ associated with its excitation blockade effect from $|\Omega_2|^2/n_1^{11/2}$. Next, the power dependence of the suppressed dip can also reflect the blockade effect and the dressing effect of $E_2$ (see Fig. 5(b2)). Initially, the saturated dressing of $E_2$ on FWM2 signal at $\Delta_1 + \Delta_2 = 0$ and gradually increased SWM1 signal are the main factors. Then the curve becomes saturated due to the blockade term ($|\Omega_2|^2/n_1^{11/2}$) at higher power level of $E_2$. As the power further increasing, the interaction between two dressing processes weakens the dressing results. Finally, one can obtain the direct blockade effect from the $P_2$ power dependence of the background as shown in Fig. 5(b3). The background is consisted of FWM2 and SWM1 signals without the dressing effect of $E_2$. The saturation is due to the blockade effect, and the descending part is due to the combination of blockade effect and dressing effect of $E_2$. Given the above descriptions and analysis of peak and dip evolution corresponding to $P_2$ strength dependence, one can deduce that the phase modulation as well as asymmetry can become more obvious by strengthening $E_2$. Meanwhile, we must note that the different principles for the increase of asymmetry are very corresponding to the three stages of power increase mentioned above. Lastly, $E_4$ power dependences in Fig. 5(c) just show the regular enhancement and suppression processes by $E_4$. The asymmetry changes are mainly aroused from the dressing effect of $E_4$.

**Discussion**

The dressed suppression and enhancement of blockade MWM processes can reveal the change in non-linear refractive index induced by cooperative atom-light interactions and corresponding dressing effects in Rydberg-EIT hot medium. On one hand, the observation of spatial shift and splitting effects of corresponding signals can visually advocate the dispersion property change of medium under blockaded effect. The transverse wave vector to explain the spatial effects is defined as
\[
\frac{\partial^n \Delta \Phi}{\partial \xi^n} = \frac{\partial^n ((2k \Delta n_L/c) \sum_{i=1}^{\infty} (-1)^i \xi^{2i}/(i!)) \cdot 2k \Delta n_L/c \sum_{i=1}^{\infty} (-1)^i \xi^{2i}}{\partial \xi^n}.
\]

The first-order differential \( \delta k_\perp = \delta \Delta \Phi_1 / \partial \xi \) can describe spatial shift/splitting effects and the second-order differential \( \partial^2 \Delta \Phi_2 / \partial \xi^2 \) can explain the focusing/defocusing effects. On the other hand, the intensity modification of the enhanced and suppressed MWM signals obtained by scanning the dressing fields, which essentially control dark and bright states, can reflect the change in refractive index of a medium for a laser or MWM signals. Further, the cooperative nonlinearity induced phase modulation can be proportional to the refractive index change caused by Rydberg energy level shift. Consequently, we can quantificationally map the phase shift by cooperative nonlinear interaction onto suppression and enhancement of MWM processes involving in Rydberg states. With the dressing asymmetry \( A_\phi \) on the modulated results defined, \( A_\Phi \propto (\Delta \Phi_{\text{WM}})/(\alpha_1 \Delta \Phi + \alpha_2 \Delta \Phi') \) is established to depict the phase shift between dressing dark and bright states, where \( \Delta \Phi \) includes the phase shifts from both Rydberg dressing states and Rydberg excitation blockade and \( \Delta \Phi' \) results from the orientations of induced dipole moments. The parameters \( \alpha_1 \) and \( \alpha_2 \) can be determined by experimental parameters such as the frequency detunings, Rabi frequencies, atom density and polarization states of laser fields.

**Methods**

**Experimental setup.** We use six light beams from three commercial external cavity diode lasers (ECDL) and one frequency-doubling laser system to couple a five-level X-type rubidium atomic system. The transition of \( D_2 \) line is driven by weak laser beam \( E_1 \) stabilized to a temperature-controlled Fabry-Perot (FP) cavity. A pair of coupling beams \( E_2 \) and \( E_2' \), also driving the transition of \( D_1 \) line for different hyperfine configuration, are from another ECDL locked to the saturated absorption signal of rubidium atom. Beam \( E_1 \), driving the Rydberg excitation is a frequency-doubled laser with high stability. We get the needed 480 nm laser \( E_2 \) by the way of frequency doubling LD2 at ~960 nm with a periodically-poled KTP crystal in an external ring resonator to generate the second harmonic wave. The strong beam \( E_1 \), adding onto the beam \( E_2 \) (in the same direction), which counter-propagates with beam \( E_0 \), drives the highly-excited Rydberg transition. \( E_4 \) and \( E_4' \) are from the same LD4. \( E_5 \) adds onto the beam \( E_1 \) by a cubic polarizing beam splitter (PBS) and \( E_5' \) propagates with \( E_4' \) symmetrically with respect to \( E_2 \). All beams are focused by two lenses (L1 and L2, respectively) with same focal length 500 mm before the cell and intersect at one point inside the cell. The 1 cm long rubidium cell is wrapped by \( \mu \)-metal and heated by the heater tape. The optical depth (OD) is 70 for atom density of \( 1.0 \times 10^{12} \) cm\(^{-3} \).

**Theoretical models for \( \Delta n \) and \( \Delta \Phi_1(U) \).** Nonlinear refractive index change is modeled by taking \( \Delta n \) as the product of the slope of the dispersion \( \partial n/\partial \omega_2 \) and the energy level shift \( (\Delta \omega_2) \) of the Rydberg state due to the Rydberg-Rydberg interaction. \( \partial n/\partial \omega_2 \) is derived from the real part of the complex susceptibility \( \chi^{(3)} \) for stationary atoms and zero-coupling detuning as

\[
\frac{\partial n_r}{\partial \omega_2} = \frac{12\pi^2}{n_0^2 c} \frac{\partial \text{Re} \left[ \chi \right]}{\partial \omega_2} = \frac{4\pi^2 N_0 \mu_0^4}{c\varepsilon_0 n_0^2 \hbar^3} \text{Re} \left[ \frac{1}{\gamma_1 \gamma_2 D_1} \left( \frac{G_2^2}{\gamma_2^2 D_1} - \frac{1}{\gamma_2} \right) \right],
\]

where \( n_0 \) is the linear refractive index and \( D_1 = \gamma_1 + |\Omega_3|/\gamma_2 + |\Omega_4|/\gamma_4 \). The energy level shift is

\[
\Delta \omega_2 = \frac{N_2}{h} \int \nu \cdot U(r - r') d^3 r',
\]

where \( U(r - r') \) is the cooperative nonlinear interaction for Rydberg atoms at \( nD \) states; \( N_2 \) is the density of excited Rydberg atoms. If we calculate the Rydberg excitation density via optical Bloch equation (OBE) by using the mean-field model2 and taking \( N_2 V_d = 1 \) & \( V_d \propto (R_d)^3 \) into account, the average Rydberg atom density \( \rho_e \) can be described as

\[
\rho_e = 3R_d^{-3}/4\pi.
\]

Here \( R_d \) is the radius of a Rydberg domain, which includes a single Rydberg atom and many ground-state atoms. By comparing with the non-blockade case, we find the following regulation as

\[
\begin{align*}
N_0^{\text{Blockade}} & \to N_0^{0.4}, \quad \Omega_2^{\text{Blockade}} \to (\Omega_2/h)^{0.4}, \\
N_2 & \approx CN_1^{0.4} \left( |\Omega_3|/h^{1.0} \right)^{0.4},
\end{align*}
\]

So the density of excited Rydberg atoms \( N_2 \) is given as

\[
N_2 \approx CN_1^{0.4} \left( |\Omega_3|/h \right)^{1.0} \Gamma_{13} + C \left( \text{Re} \left[ \gamma_1 \right] \right)^{1/2} \left( \text{Im} \left[ \gamma_1 \right] \right)^{1/2}.
\]

Here \( \Gamma_{13} = \Gamma_{13} + i\Delta_{13} \) and \( C \) is a constant mainly determined by the coefficient of Rydberg-Rydberg
interaction and resulting from numerical integration outside the given sphere and the atom excitation
efficiency between |0\rangle and |1\rangle. Therefore, the change in refractive index can be defined as
\[
\Delta n_r = \frac{\partial n_r}{\partial \omega^2} \Delta \omega = \frac{4\pi^2 N_d d_{10}^2 N_2}{\epsilon_0 n_0^2 c^2 \hbar^4} \text{Re} \left[ \frac{1}{\gamma_1 \gamma_2 D_1} \left( \frac{|\Omega_2|^2}{\gamma_2^2} - \frac{1}{\gamma_2} \right) \right] \int v_r U(r - r') d^3r'.
\] (15)

The induced phase modulation under the cooperative nonlinear interaction is
\[
\Delta \Phi_1(U) = L\omega^2 \Delta n_r/c = \frac{4\pi^2 N_d d_{10}^2 |\Omega_2|^2 N_2}{\epsilon_0 n_0^2 c^2 \hbar^4} \text{Re} \left[ \frac{1}{\gamma_1 \gamma_2 D_1} \left( \frac{|\Omega_2|^2}{\gamma_2^2} - \frac{1}{\gamma_2} \right) \right] \int v_r U(r - r') d^3r'.
\] (16)

References
1. Mohapatra, K., Bason, M. G., Butscher, B., Weatherill, K. J. & Adams, C. S. A giant electro-optic effect using polarizable dark states. Nature Phys. 4, 890–894 (2008).
2. Tong, D. et al. Local blockade of Rydberg excitation in an ultracold gas. Phys. Rev. Lett. 93, 063001 (2004).
3. Singer, K., Reetz-Lamour, M., Amthor, T., Marcassa, L. G. & Weidemüller, M. Suppression of excitation and spectral broadening induced by interactions in a cold gas of Rydberg atoms. Phys. Rev. Lett. 93, 163001 (2004).
4. Vogt, T. et al. Dipole blockade at Förster resonances in high resolution laser excitation of Rydberg states of cesium atoms. Phys. Rev. Lett. 97, 083003 (2006).
5. Pritchard, J. D., Weatherill, K. J. & Adams, C. S. Nonlinear optics using cold Rydberg atoms. Annual Rev. of Cold At. & Mol. 1, 301–350 (2013).
6. Brekke, E., Day, J. O. & Walker, T. G. Four-wave mixing in ultracold atoms using intermediate Rydberg states. Phys. Rev. A. 78, 063830 (2008).
7. Sevinçli, S., Henkel, N., Ates, C. & Pohl, T. Nonlocal nonlinear optics in cold Rydberg gases. Phys. Rev. Lett. 107, 153001 (2011).
8. Rolle, A., Eppli, G., Kohler, H., Low, R., & Pfau, T. Four-wave mixing involving Rydberg states in thermal vapor. Phys. Rev. A. 85, 063821 (2012).
9. Thompson, D. C., Weinfurter, E., Xu, G.-X. & Stoicheff, B. P. Frequency shifts and line broadenings in collisions between Rydberg and ground-state alkali-metal atoms. Phys. Rev. A. 35, 690–700 (1987).
10. Vitrant, G., Raimond, J. M., Gross, M. & Haroche, S. Rydberg to plasma evolution in a dense gas of very excited atoms. J. Phys. B. 15, L49–L55 (1982).
11. Robinson, M. P., Tolra, B. L., Noel, M. W., Gallagher, T. F. & Pillet, P. Spontaneous evolution of Rydberg atoms into an ultracold plasma. Phys. Rev. Lett. 85, 4466–4469 (2000).
12. Lukin, M. D., Matsko, A. B., Fleischhauer, M. & Scully, M. O. Quantum noise and correlations in resonantly enhanced wave mixing based on atomic coherence. Phys. Rev. Lett. 82, 1847–1850 (1999).
13. Lukin, M. D., Hemmer, P. R. & Scully, M. O. Resonant nonlinear optics in phase-coherent media. Adv. At. Mol. Opt. Phys. 42, 347–386 (2000).
14. Xiao, M., Li, Y. Q., Jin, S. Z. & Gea-Banacloche, J. Measurement of dispersive properties of electromagnetically induced transparency in rubidium atoms. Phys. Rev. Lett. 74, 666–669 (1995).
15. Gea-Banacloche, J., Li, Y. Q., Jin, S. Z. & Xiao, M. Electromagnetically induced transparency in ladder-type inhomogeneously broadened media: Theory and experiment. Phys. Rev. A. 51, 576–584 (1995).
16. Baluktsian, T., Huber, B., Low, R. & Pfau, T. Evidence for strong van der Waals type Rydberg-Rydberg interaction in a thermal vapor. Phys. Rev. Lett. 110, 123001 (2013).
17. Carr, C. et al. Three-photon electromagnetically induced transparency using Rydberg states. Opt. Lett. 37, 3858–3860 (2012).
18. Li, P. et al. Phase control of bright and dark states in four-wave mixing and fluorescence channels. Appl. Phys. Lett. 101, 081107 (2012).
19. Carr, C. Ritter, R. Wade, C. G. Adams, C. S. & Weatherill, K. J. Nonequilibrium phase transition in a dilute Rydberg ensemble. Phys. Rev. Lett. 111, 113901 (2013).
20. Pritchard, J. D. et al. Cooperative atom-light interaction in a blockaded Rydberg ensemble. Phys. Rev. Lett. 105, 193603 (2010).
21. Wang, H., Goorskey, D. & Xiao, M. Enhanced kerr nonlinearity via atomic coherence in a three-level atomic system. Phys. Rev. Lett. 87, 073601 (2001).
22. Zheng, H. et al. Blockaded six- and eight-wave mixing processes tailored by electromagnetically induced transparency scissors. Laser Phys. 24, 045404 (2014).
23. Petch, J. C., Keitel, C. H., Knight, P. L. & Marangos, J. P. Role of electromagnetically induced transparency in resonant four-wave-mixing schemes. Phys. Rev. A. 53, 543–561 (1996).
24. Zhang, Y., Brown, A. W. & Xiao, M. Opening four-wave mixing and six-wave mixing channels via dual electromagnetically induced transparency windows. Phys. Rev. Lett. 99, 123603 (2007).
25. Zhang, L., Feng, Z., Zhao, J., Li, C. & Jia, S. Evolution of the pairs of ultracold Rydberg atoms in the repulsive potential. Opt. Express. 18, 11599–11606 (2010).
26. Sun, J. et al. Comparison of two two-photon dressed rules in multi-wave mixing. IEEE Photon. J. 6, 6100109 (2014).
27. Li, W. H., Mouzachos, I., Noel, M. W. & Gallagher, T. F. Millimeter-wave spectroscopy of cold Rb Rydberg atoms in a magneto-optical trap: Quantum defects of the ns, np, and nd series. Phys. Rev. A. 67, 052502 (2003).
28. Li, Y. & Xiao, M. Transient properties of an electromagnetically induced transparency in three-level atoms. Opt. Lett. 20, 1489–1491 (1995).

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Author Contributions
Z.Y.Z. and H.B.Z. wrote the main manuscript and contributed to the theoretical and experimental analysis. Y.P.Z. and M.X. provided the idea. Y.L.T., X.X.W. and D.Y.Z. contributed to the presentation and execution of the work. All authors discussed the results and contributed to the writing of the manuscript.
Additional Information

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