Novel Rydberg eight-wave mixing process controlled in the nonlinear phase of a circularly polarized field

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Abstract: Eight-wave mixing (EWM) is a seven-order nonlinear process that can reflect nonclassical features within multiple optical fields, thus imparting certain advantages. In this study, we directly observed the EWM spectrum and spatial images that show Rydberg atoms under a circularly polarized probe field in a five-level coherently prepared atomic system. Such circular polarization dressing fields can obtain high-contrast Rydberg EWM overcome the difficulties of several multi-wave mixing (MWM) signals always coexist, and the multi-parameter controlling Rydberg EWM mechanism is established by changing the power and detuning and polarization of the dressing fields. These controllable high-order MWM processes present a contrast ratio of 96% and a narrow linewidth of <30 MHz compared with low-order mixing processes under identical conditions (e.g., six-wave mixing). The corresponding MWM spatial images are presented, and they can partly reflect the underlying nonlinear phase variation, whereas the given theory can predict the experimental results.

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References and links

Nonlinear polarization is an important physics quantity that reflects the interaction between optics and media, and it can be expanded in series as $P(t) = \epsilon_0 \chi^{(1)} E(t) + \chi^{(3)} E^3(t) + \chi^{(5)} E^5(t) + \chi^{(7)} E^7(t) + \ldots$ in an isotropic medium. Multi-wave mixing (MWM) represents different odd-order nonlinearity polarizations $\chi^{(i)}$. For example, four-wave mixing (FWM) is represented by $\chi^{(3)}$ and eight-wave mixing (EWM) is represented by $\chi^{(7)}$ and is produced by seven light fields; it can be used to transfer information on the light field to the medium. Such MWM processes can make the energy transfer between different waves [1, 2], can be applied for atom quantum memories [3] and unique wavelength creation [4], and can be correlated with many photon pairs [5]. Along different lines, highly excited Rydberg atoms, owing to their intriguing properties such as long lifetime, large dipole moment and strong interactions at long distances have been extensively investigated for quantum information applications [6]. However, many studies related to Rydberg atoms have primarily been conducted in magneto-optical traps [7, 8], which dramatically limits the practical applications of these atoms because of the complexity of the system. In recent years, Rydberg atoms have been investigated in thermal vapor using the electromagnetically induced transparency (EIT) technique, which has attracted the attention of many researchers to demonstrate non-destructive detection [9], GHz Rabi-flopping [10], Rydberg-Rydberg interaction [11], and have direct application on microwave...
sensing [12]. Because EIT is a quantum coherent effect that has dual functions, including its depiction by the imaginary density matrix element [13], which can theoretically represent absorption and provide a relatively ideal environment to suppress the Doppler Effect. The other function is the real part of the density matrix element, which describes dispersion; therefore, EIT can be used as a tool to apply for slow-light group velocity matching and enhance the intensity of MWM [14]. Consequently, Rydberg MWM processes can be investigated using the dual functions of EIT. In addition, inspired by the demonstration of Rydberg four-wave mixing (FWM) in a degenerate-energy-level system in thermal vapour, which revives signals beyond the frozen-gas limit [15, 16], and six-wave mixing (SWM) is used to convert millimeter waves to an optical field [17]. Our group has generated co-occurring SWM and EWM in a non-degenerate Rydberg thermal atomic ensemble [18, 19]. For these two MWM processes, EWM presents obvious advantages because of the multiple EIT windows. For example, EWM has a much narrower linewidth than SWM and can possess certain advantages in microwave sensing [20]. However, directly extracting and controlling EWM is always difficult. In this paper, this problem will be solved and the corresponding parametric control mechanism is established in circularly polarized probe field. Meanwhile, the spatial images of MWM are obtained and analyzed in these processes to study the nonlinear phase variation are observed.

In this letter, we experimentally present the nonlinear spectra and spatial images showing the polarized-dressing Rydberg EWM process, which is enhanced by co-occurring EIT windows in a five-level atomic system. Quarter-wave plate (QWP) are used to change the polarizations of the probe field to directly observe the intensity evolution of Rydberg (37D5/2) windows in a five-level atomic system. For these two MWM processes, EWM presents obvious advantages because of the multiple EIT windows. For example, EWM has a much narrower linewidth than SWM and can possess certain advantages in microwave sensing [20]. However, directly extracting and controlling EWM is always difficult. In this paper, this problem will be solved and the corresponding parametric control mechanism is established in circularly polarized probe field. Meanwhile, the spatial images of MWM are obtained and analyzed in these processes to study the nonlinear phase variation are observed.

2. Experimental scheme

The experiment was implemented in a thermal 85Rb vapour cell where ground atomic density is about 1.0 × 10^{12} cm^{-3}. The 1cm long cell with natural abundance Rb vapour is wrapped with μ-metal sheets and heated to 65°C. Five laser beams from four commercial external cavity diode lasers (ECDLs) were used to establish the energy-level structure system in Fig. 1(a). Firstly, the weak probe beam \( E_1 \) (with a wavelength of 780 nm, power 1mw, and a diameter of 0.8 mm) drives 5S1/2(F = 3) to 5P3/2(F = 3). Beams \( E_5 \) (780 nm, 10mw, 1 mm) and \( E'_5 \) (780 nm, 23mw, 1 mm) derived from the same ECDL split through a polarization beam splitter (PBS) that connects the transition between 5S1/2(F = 3) and 5S1/2(F = 2). With these three beams injected, one FWM that satisfies the phase-matching condition \( k_f = k_1 + k_3-k'_4 \) can be generated in the \( \Lambda \)-type three-level subsystem \( \left| 0 \right> \leftrightarrow \left| 1 \right> \leftrightarrow \left| 3 \right> \). Then the high-power coupling beam \( E_3 \) (480 nm, 100mw, 1 mm) produced by frequency doubling a 960nm ECDL is added to drive 5P3/2(F = 3) to the 37D5/2 Rydberg state. As a result, one EIT-assisted SWM \( E_{S1} \) (satisfying phase-matching condition \( k_{S1} = k_1 + k_3-k'_4 + k_2-k_3 \)) can occur. The EIT window for SWM is generated by \( \left| 0 \right> \leftrightarrow \left| 1 \right> \leftrightarrow \left| 2 \right> \) transition. In the following, the dressing field \( E_4 \) (776 nm, 35mw, 0.9 mm) participates in driving the transition of 5P3/2(F = 3) to \( 5D_{3/2}(F = 2) \) to generate the other SWM \( E_{S2} \) assisted by \( \left| 0 \right> \leftrightarrow \left| 1 \right> \leftrightarrow \left| 4 \right> \) EIT window (satisfying phase matching condition \( k_{S2} = k_1 + k_3-k'_5 + k_4-k_5 \)) in the \( \left| 0 \right> \leftrightarrow \left| 1 \right> \leftrightarrow \left| 3 \right> \leftrightarrow \left| 4 \right> \leftrightarrow \left| 1 \right> \leftrightarrow \left| 2 \right> \leftrightarrow \left| 1 \right> \) transition. At last, an EWM process \( k_{EWM} = k_1 + k_3-k'_5 + k_2-k_3 + k_4-k_5 \) with two overlapped EIT windows should be obtained in the process \( \left| 0 \right> \leftrightarrow \left| 1 \right> \leftrightarrow \left| 3 \right> \leftrightarrow \left| 4 \right> \leftrightarrow \left| 1 \right> \leftrightarrow \left| 2 \right> \leftrightarrow \left| 1 \right> \), where both \( k_3 \) and \( k_4 \) are used twice in the same propagation direction.
Fig. 1. (colour online) (a) K-type five-level system of 85Rb atoms, where $F = 3$ (|0⟩) and $F = 2$ (|3⟩), a low-lying excited state $5S_{1/2}$, $F = 3$ (|1⟩), a low-lying excited state $5P_{3/2}$, $F = 2$ (|4⟩), and a highly excited Rydberg state $37D_{5/2}$ (|2⟩) are involved. (b) Spatial arrangement of laser beams in the experiment and EWM phase matching condition diagram, where $E_i$ on behalf of different laser field, $E_{M}$ is the MWM field. (c) Zeeman sublevels with various transition pathways.

All laser beams were spatially aligned as shown in Fig. 1(b). The probe beam $E_1$ (frequency $\omega_1$, wave vector $k_1$, Rabi-frequency $\Omega_1$ and frequency detuning $\Delta_1$) can be modulated by a QWP before entering the atomic medium. Here, $\Delta_i = \omega_i - \omega_j$ is the frequency detuning, $\omega_i$ is the frequency of laser $E_i$, $\omega_j$ is the resonant transition frequency between $|i⟩$ and $|j⟩$, $\Omega_i$ is the Rabi frequency that corresponds to optical field $E_i$ and can be described as $\mu_i E_i / \hbar$, $\mu_i$ is the transition dipole moment, and $E_i$ is the energy of the laser field. Two pump beams $E_3$ ($\omega_3$, $k_3$, $\Omega_3$, $\Delta_3$) and $E_3'$ ($k'_3$, $\Omega'_3$, $\Delta'_3$) with an angle of 0.3° between them propagate in the opposite direction of $E_1$. Two dressing fields $E_2$ ($\omega_2$, $k_2$, $\Omega_2$, $\Delta_2$) and $E_4$ ($\omega_4$, $k_4$, $\Omega_4$, $\Delta_4$) are injected into the cell in the opposite direction of $E_1$ and overlap in the central point of the sample. These MWM signals all emit in the opposite direction of $E_3'$ (see Fig. 1(b)) and are identified by their corresponding EIT windows and detected by an avalanche photodiode detector (APD). The generated MWM signals are extracted through a PBS cube, the horizontal component of the generated MWM signals is detected by the APD, and the vertical component is received by a charge coupled device (CCD) camera. The probe transmission is detected by the other APD. When QWP is changed, the polarization of probe field will vary from linear polarization to circular one so that the selection rule and CG coefficient are also modified. Zeeman sublevels with various transition pathways and corresponding Clebsch-Gordan (CG) coefficient are shown in Fig. 1(c).

3. Basic theory

Theoretically, the imaginary part of the first-order density matrix element $\rho_{01,01}^{(1)}$ proportionally determines the absorption of the probe beam and intensity of the probe transmission signal. The corresponding matrix elements can be obtained by solving the coupled density-matrix equations [21] and density-matrix element $\rho_{01,01}^{(1)}$ with two EIT processes, which is described as follows:

$$\rho_{01,01}^{(1)} = i\Omega_{01}/(d_{01})^2 + \Omega_{11}/(d_{11})^2 + \Omega_{22}/(d_{22})^2 + \Omega_{33}/(d_{33})^2 + \Omega_{44}/(d_{44})^2,$$  \hspace{1cm} (1)
where \( d_{\nu r, u} = \Gamma_{\nu r, u} \phi_u + i \Delta_u \); \( d_{\nu r, v} = \Gamma_{\nu r, v} \phi_v + i (\Delta_v - \Delta_u) \); \( d_{\nu r, w} = \Gamma_{\nu r, w} \phi_w + i (\Delta_w + \Delta_v) \); \( \Gamma_{ij} \) is the Rabi frequency of each optical field considering Zeeman sublevel in which \( q = 0, +1 \) and \(-1\), \( M \) is the Zeeman sublevel of the polarized probe field; and \( \Gamma_y \) is the transverse relaxation rate for states \(|r\rangle\leftrightarrow|f\rangle\), \( n \) is the principle quantum number.

Generally, the expression of density matrix elements that correspond to MWM signals can also be depicted by solving the density matrix equations [21]. Because the polarization of the probe field for the classical FWM signal is changed, the third-order density matrix element can be obtained via pathway \( \rho_{\nu r, u}^{(0)} \rightarrow \rho_{\nu r, v}^{(1)} \rightarrow \rho_{2 \nu r, u}^{(2)} \rightarrow \rho_{2 \nu r, v}^{(3)} \); 

\[
\rho_{\nu r, v}^{(3)} = -i \Omega_{\nu r, u}^{0, \phi} \left| \Omega_{\nu r, v}^{0, \phi} \right|^2 e^{i \phi} / (d_{\nu r, u} + \Omega_{\nu r, u}^{0, \phi} / \Gamma_{0 u, v})^2 \Delta_{3 \nu r, v}.
\] (2)

The amplitude of \( \rho_{\nu r, v}^{(3)} \) determines the intensity of the FWM. In Eq. (2), the phase factor \( \phi \) is equal to \( \phi_{i} + \phi_{r} + \phi_{v} \), where \( \phi_{i} \) characterizes the nonlinear phase shift induced by strong field \( E_{i} (E'_{i}) \). \( 0, + \) and \(- \) represent linear polarization, right circular polarization and left circular polarization of optical field, respectively. For nonlinear phase shift induced by strong field can be depicted as \( \phi = 2k_{p,r,s} n_{0} I_{r} e^{-c^{-2} r^2 / \Omega_{0, p}^{s}} / (n_{0} I_{p,r,s}) \) according to nonlinear Schrödinger equation that describe propagation of probe and MWM. In the expression of \( \phi \), \( \zeta \) is the centre coordinate of \( E_{i} \) in the transverse dimension relative to the centre coordinate of \( E_{1} \) as the original point; nonlinear refractive index \( n_{r} = \text{Re} \rho_{r, p}^{(3)} / (c n_{0}) \) with set \( n_{0} \) as the linear refractive index; \( k_{p,r,s} \) and \( I_{p,r,s} \) are the corresponding wave vector and intensity of the probe transmission, FWM, SWM and EWM, respectively; Phase \( \phi_{i} + \phi_{r} + \phi_{v} \) is the nonlinear phase represented by polarized \( E_{i} \) fields, which can be called the polarized nonlinearity phase. When the QWP changes the polarization of \( E_{i} \) from linear to circular, the nonlinear phase \( \phi_{i} + \phi_{r} + \phi_{v} \) will be generated, and \( E_{1} \) is decomposed into balanced left- and right-circularly polarized beams. During the interaction between the circularly polarized field and the strong pump field, various transitions occur among Zeeman sublevels to generate different MWM signals. Different transition paths are associated with CG coefficients can determine the intensity of the MWM signals due to CG coefficients make the Rabi-frequency \( \Omega_{r} \) altered with different polarizations of light. Figure 1(c) shows the transition path and CG coefficients at different polarization states of the probe beam. The circularly polarized probe field can generate a circularly polarized MWM and cause a phase difference between left-circular polarization and right-circular polarization. The difference of the nonlinear phases is

\[
\Delta \phi = \frac{\alpha_{i}}{c} \int_{L} \left[ \chi(\nabla) - \chi^{(3)} \right] |E(\alpha_{i})|^2 dL
\]

which are obtained via optical field transmission couple wave equation, where \( \chi^{(3)} \) and \( \chi^{(3)} \) are third-order nonlinear susceptibility caused by right-circularly polarized and left-circularly polarized, respectively, \( L \) is the length of sample. In detail, the third-order nonlinear susceptibility caused by right-circularly polarized are described as \( \chi^{(3)} = N \mu_{10}^{3} / h \epsilon_{0} \Omega_{r} \rho_{10}^{(3)} \), where \( \Omega_{r} \) is the Rabi-frequency of MWM, \( N \) is atomic density, \( \rho_{10}^{(3)} = \sum_{M} (\Gamma_{1 u, v} + i \Delta_{u}) (\Gamma_{2 u, w} + i \Delta_{w}) \). Correspondingly, the left-circularly polarized case has \( \chi^{(3)} = N \mu_{10}^{3} / h \epsilon_{0} \Omega_{l} \rho_{10}^{(3)} \),
\[
P_{0}^{(3)} = \sum_{\mu} \frac{i \Omega_{\mu}^{\mu} |\Omega_{2,\mu+1}^{\mu}|^2}{(\Gamma_{1,\mu+1} + i \Delta_1)^2 (\Gamma_{2,\mu+1} + i (\Delta_1 - \Delta_2))}.
\]

Considering that the generated MWM signals are extracted through a PBS cube, the horizontal component is detected by an APD and can be described as
\[
I_B = I_M \cos^2(\theta + \Delta \phi) = (I_{c+} + I_{c-}) \cos^2(\theta + \Delta \phi),
\]
where \(\theta\) is the angle of wave plate, \(I_{c+}\) and \(I_{c-}\) are the right-circularly and left-circularly polarized intensity of \(E_1\), respectively. The vertical component of the generated MWM signals which is described as
\[
I_V = I_M \sin^2(\theta) (\theta - \Delta \phi) = + \Delta \phi
\]
is then injected into a charge-coupled device (CCD) camera, where \(I_{c+}\) and \(I_{c-}\) are horizontal polarization of MWM and vertical polarization component of MWM, respectively.

For Rydberg interaction, we have calculated the Rydberg excitation density via the optical Bloch equation (OBE) using the mean-field model and obtain corresponding excitation density \(N_2 = C N_1 \left( | \Omega_{2u}^{\mu} / n_1 |^{0.4} \right)\), their detailed deduction process can be found in [22], where \(N_1\) is the density of atoms at level \(|1\rangle\) (with the EIT and optical pumping effect considered) and given by
\[
N_1 = \frac{1}{2} N_0 \left( \text{Re}[d_{3u} + |\Omega_{3u}^{\mu}|^2 / d_{3u} + |\Omega_{2u}^{\mu}|^2 / d_{2u}] \right)
\]
given by
\[
d_{3u} = \Gamma_{1u} + i \Delta_1; N_0 \text{ is the density of ground-state atoms; and } C \text{ is a constant mainly determined by the coefficient of the vdW interaction and results from the numerical integration outside the given sphere and excitation efficiency between } |0\rangle \text{ and } |1\rangle. \]

We utilized these calculation results to modify the fifth-order density matrix element which determine the SWM signals considering Rydberg interaction, meanwhile via perturbation chains
\[
\rho_{u,0}^{(0)} \rightarrow \rho_{u,0}^{(1)} \rightarrow \rho_{u,0}^{(2)} \rightarrow \rho_{u,0}^{(3)} \rightarrow \rho_{u,0}^{(4)} \rightarrow \rho_{u,0}^{(5)}
\]
such fifth-order density matrix element can be depicted as
\[
\rho_{u,0}^{(5)} = -i \Omega_{u,0}^{\mu} \left[ \Omega_{3u,0}^{\mu} \right] \left[ \Omega_{2u,0}^{\mu} \right] e^{i(\Delta \phi_1 + \phi)} / n_1 m \left[ d_{3u} + 1 / d_{2u} \right]
\]
where \(m = d_{3u} + |\Omega_{3u}^{\mu}|^2 / d_{3u} + |\Omega_{2u}^{\mu}|^2 / d_{2u}\). Here, \(\Delta \phi_1(U)\) is the phase modulation caused by Rydberg-Rydberg interaction, due to the van der Waals interaction, the change in refractive index \(n_1\) of a medium for a probe laser (or MWM signal) frequency \(\omega_1\) is
\[
\Delta n = (\partial n / \partial \omega_1) \Delta \omega_1,
\]
where \(\partial n / \partial \omega_1 = (n_1 - 1) / \omega_1\) and \(n_1\) is the group refractive index. The principal quantum number \(n\) dependence of the Kerr coefficient for constant coupling laser power is modeled by taking \(\Delta n\) as the product of the slop of the dispersion \(\partial n / \partial \omega_1\) and the energy level shift \(\Delta \omega_2\) of the Rydberg state due to the van der Waals interaction. Utilizing the real part of the complex susceptibility \(\chi\) for stationary atoms and zero-coupling detuning, \(\partial n / \partial \omega_2\) can be described as
\[
\frac{\partial \text{Re}[\chi]}{\partial \omega_2} = 3 \pi N_0 \lambda_1 \Gamma_{1u} \frac{4 \Omega_{2u}^{\mu}}{16 \Gamma_{10}^2 \Gamma_2 + 8 \Gamma_{10} \Gamma_2 \Omega_{2u}^{\mu} + \Omega_{2u}^{\mu}}.
\]

where \(\lambda_1\) is the probe (MWM) field wavelength. The energy level shift caused by van der Waals interaction is
\[
\Delta \omega_2 = \frac{N_2}{\hbar} \int \rho U(r - r') d^3 r'
\]
By defining $\Gamma = \sqrt{\Gamma_1 \Gamma_2}$, the change in refractive index can be got as

$$
\Delta n = -12 \pi N_2^2 N_0 \frac{\Omega^2_{2\leftrightarrow r}}{1641 + 84 \Gamma_{2\leftrightarrow r}^2} \int_{r'} U(r-r') d^3r'.
$$

(6)

So the phase modulation under the interaction can be expressed as follows:

$$
\Delta \Phi_g (U) = L \omega_2 \Delta n / c = \frac{4 \pi^2 N_e^4 n_0^4 L \omega_2 N_e}{\epsilon_0 n_0^2 c^2 h^4} \text{Re} \left( \left| \Omega_{2\leftrightarrow r} \right|^2 \right) \int_{r'} U(r-r') d^3r',
$$

(7)

where $D = d_{1\leftrightarrow r} + \Omega_{2\leftrightarrow r} / d_{2\leftrightarrow r} + \Omega_{4\leftrightarrow r} / d_{4\leftrightarrow r}$; $U(r-r') \propto C_6 / R^6$ [23] is the van der Waals (vdW) interaction between Rb atoms at nD states; $n_0$ is the linear refractive index; $\Delta n$ is the variation of the refractive index; and $N_2$ is the density of excited Rydberg atoms. $\Delta \Phi_g (U)$ can directly affect the intensity of Rydberg signals. Similarly, by considering the dressing effects of the $E_1$, $E_2$ and $E_4$ beams, the EWM process can be determined based on the corresponding seventh-order density matrix element:

$$
\rho^{(7)}_{2\leftrightarrow r} = i \Omega^{2\leftrightarrow r} \left( \left| \Omega_{2\leftrightarrow r} \right|^2 / \left| \Omega_{4\leftrightarrow r} \right|^2 \right) / \left( \left| \Omega_{1\leftrightarrow r} \right|^2 / \left| \Omega_{2\leftrightarrow r} \right|^2 \right) e^{i \left( \Delta \Phi_4 + \phi_{2\leftrightarrow r} \right)} / m d_{1\leftrightarrow r}^2 d_{2\leftrightarrow r}^2 d_{4\leftrightarrow r}. 
$$

(8)

4. Results and Discussion

First, the high-contrast $37D_{5/2}$ Rydberg EWM signal is observed with a circularly polarized probe field. The MWM spectrum is the strongest with the detuning fixed at $\Delta_1 = -400$ MHz. Figures 2(a) and 2(b) show the probe transmission signals and corresponding MWM signals versus $\Delta_2$ at different rotation angles $\theta$ of the QWP with $E_4$ on and off, respectively. The values in the horizontal axis in Fig. 2 denote the values of $\theta$. Figures 2(a1) and 2(a2) are the cases with all beams on, and each peak in Fig. 2(a1) shows the probe transmission signals (at $\Delta_2 \approx -\Delta_1$) induced by the dressing field $E_2$ according to Eq. (1), which determines the intensity of the probe transmission. The EIT window caused by $E_4$ (hiding in the baseline) can interact with the EIT of $E_2$. The heights of the EIT peaks are maximum at $\theta = 0$, gradually reach the lowest point at $\theta = 45^\circ$, and finally return to the original height when $\theta = 90^\circ$. This phenomenon can be explained by Eq. (1) because different CG coefficients determine the different transitions between Zeeman sublevels, which can lead to different Rabi frequencies.

For example, $\left[ \Omega_{1\leftrightarrow r}^2 \right] / \left[ \Omega_{2\leftrightarrow r}^2 \right] = 5$ in Fig. 1(c) illustrates that the dressing effects of $\Omega_{1\leftrightarrow r}^2$ are greater than that of $\Omega_{2\leftrightarrow r}^2$, and factor $\Omega_{1\leftrightarrow r}^2 / \Gamma_{1\leftrightarrow r} \rho_{1\leftrightarrow r}$ also plays an important role such that the height of the EIT peaks are lowest at $\theta = 45^\circ$. The dashed line in Fig. 2(a1) represents the transparent degree of $E_4$ obtained by the actual experiment offset, which has the same evolution regularity as the peaks. Figure 2(b1) shows the identical probe transmission spectra as Fig. 2(a1) except for blocking $E_4$, where the peaks of EIT are created by $E_2$ and interactions do not occur with the EIT of $E_4$. By comparing Figs. 2(b1) with 2(a1), the peaks in Fig. 2(a1) are smaller than those in Fig. 2(b1) at the identical polarization of $E_1$, which is mainly caused by the cascade relation between the dressing effect of $E_2$ and $E_4$, which is represented by the respective terms $\left| \Omega_{2\leftrightarrow r} \right|^2 / \left| \Omega_{4\leftrightarrow r} \right|^2$ and $\left| \Omega_{2\leftrightarrow r} \right|^2 / \left| \Omega_{4\leftrightarrow r} \right|^2$ in Eq. (1).

The corresponding MWM signals with and without $E_4$ are shown in Figs. 2(a2) and 2(b2), respectively. Because the frequency of the $E_2$ field is scanned, the FWM signal is in the baseline of observed signals. Each peak in Fig. 2(a2) is a Rydberg MWM signal, which consists of Rydberg SWM and EWM, whereas Fig. 2(b2) shows the pure Rydberg SWM signal. An analysis and comparison of Figs. 2(a2) and 2(b2) shows that the MWM (SWM +
EWM) peak is smaller than SWM at $\theta = 0^\circ$ and $90^\circ$. However, the suppression dip of the pure SWM appears at $\theta = 45^\circ$, whereas the MWM (SWM + EWM) signal shows a peak.

In other words, the high-contrast EWM signal can be observed in a circularly polarized probe field and suppressed SWM occur in the linearly polarized case, which represents an interesting phenomenon that can help us to directly extract EWM from mixed MWM signals. The contrast ratio of EWM compared with other MWM (FWM + SWM in the baseline) is approximately 96%, which is obtained based on the difference between the highest point of the MWM spectrum and the baseline (FWM + SWM). Such an EWM is generated by a seven-photon process assisted by two co-occurring EIT windows. The linewidth of the EWM (<30 MHz) can be described by $\Delta \omega = \Delta \omega_{1}(\Delta \omega_{2}/\Delta \nu_{2})$, where $\Delta \omega_{ij}$ is the linewidth of the EIT window (the measured linewidth of the Rydberg EIT is approximately 30 MHz in the experiment [18]). $\Delta \nu_{ij}$ suppresses the absorption of the field that excites the transition $|i\rangle \leftrightarrow |j\rangle$, and $\Delta \nu_{ij}$ is the linewidth of the absorption spectrum without the dressing effect. According to this description formula, one can see more EIT windows render the linewidth of Rydberg EWM signals much narrower. The direct observation of the EWM can be attributed to the stronger dressing effect from the circularly polarized $E_1$ according to Eq. (3). When $\theta = 45^\circ$, the Rydberg SWM is suppressed to generate a dip, and the term $\left | \Omega_{10}^{\pm} / \Gamma_{01}^{\pm} \right |$ behaves as a constant in Eq. (3). Although EWM is also suppressed by $\left | \Omega_{21}^{\pm} / \Gamma_{12}^{\pm} \right |$ as shown in Eq. (8), the peak of EWM remains observable in the experiment.

When $E_1$ is linearly polarized, the dressing effect of $E_1$ is weaker; therefore, the suppression effect of $E_4$ is easily reflected, and only suppressed SWM signals are observed. To sufficiently certify this process, we present Figs. 2(c) and 2(d) to show the probe transmission signals and SWM signals under circularly polarized $E_1$ change with power of $P_1$ and $P_2$, respectively. With a fixed value of $P_2 = 100$ mW and increasing $P_1$, the SWM changes from a small peak to a dip and the depth of the dip increases. When $P_1$ is small, the term $\Omega_{10}^{\pm} / \Gamma_{01}^{\pm}$ is also sufficiently small; therefore, the peaks of SWM initially increase. When
Further increases, the term $\Omega_{\text{SWM}}^1 / \Gamma_{\text{SWM},\nu}$ begins to suppress SWM. When $P_1 = 0.5$ mW and $P_2$ changes and the suppression dip for SWM varies, which further demonstrates that the suppression dip is mainly caused by $E_1$.

We also detect images of the probe and MWM with the same condition in Fig. 2(b) to visually investigate the nonlinear dispersion property that Rydberg excitation induces. Rydberg EIT can cause modifications on the refractive index and nonlinear phase shift due to the interparticle interactions in the nonlinear processes [26]. Nonlinear dispersion related to spatial location can cause spatial focusing (or defocusing), shift, and splitting [27] so that the spatial effect of corresponding images can visually advocate the change of nonlinear dispersion. Details for the theoretical derivation of $\Delta n$ and $\Delta \Phi$ have been discussed in basic theory part. The spatial focusing, splitting and shift phenomena influenced by $\Delta \Phi$ can be used to analyze the nonlinear dispersion property induced by Rydberg excitation. Figures 2(e) and 2(f) show the probe and MWM images versus $\Delta_2$ with the angle of the QWP at 0°, 35°, 45°, 55° and 65°, respectively. The images at these five certain $\theta$ values can demonstrate the evolution of the MWM spatial transmission well. The case of $\theta = 90°$ is almost same with $\theta = 0°$. From the shape evolution of probe images as shown in Fig. 2(e), the spatial variation phenomena cannot be observed in these results, which illustrates that the probe transmission images cannot reflect the spatial characters. The SWM images that correspond to Fig. 2(e) are shown in Fig. 2(f). In these figures, different types of images are obtained with different polarized probe fields, which demonstrate that different types of images are caused primarily by the polarized nonlinear phase $\phi_{\text{non}}$ in Eq. (3). When the polarization of the probe field is near the circular polarization as shown in Fig. 2(f2), the spatial splitting in the multi-photon coherent SWM images is observed in the $y$ direction; i.e., the polarized probe field overlaps with vertically polarized $E_2$ in the $y$ direction. When the scanning of $E_2$ approaches 0 MHz, the energy level of Rydberg begins to play a role and make the image focusing in Fig. 2(f4), which is consistent with the theory that the attractive atomic interaction causes self-focusing nonlinearities reported in [24]. In particular, interesting vortex-like images are observed in Fig. 2(f5) because $E_1$, $E_3$, and $E_3'$ are superposed in a certain polarized field match and the spatial patterns are formed. This spatial pattern is caused by the multi-beam superposition, which makes the spatial polygon patterns form and induces a new phase factor [25]. The image pattern looks like a circular shape, which can be understood by the phase factor $e^{i\phi}$ in Eq. (3). In Fig. 2(f5), when the scanning frequency of $E_2$ approaches resonance, the Rydberg interaction can make $\Delta \Phi$ vary to change $\exp(i\Delta \Phi)$, and the vortex-like image gradually becomes focused because the factor $\exp[i(\Delta \Phi + \varphi)]$ in Eq. (3) varies. Consequently, the evolution of the SWM images can be attributed to two aspects of the nonlinear phase: the polarized nonlinear phase changes the SWM transmission image style and the Rydberg interaction makes the SWM signal focus.

Figure 3 shows the evolution of the Rydberg EWM signal as a function of the power of the external field $E_4$ at different powers of the probe field with a circular polarization. By analysing Fig. 2(c2), we set the power of $E_1$ to 100 $\mu$W, 500 $\mu$W and 1 mW, which makes the Rydberg SWM exhibit a peak, a half dip and a half peak, and a dip, respectively. When the power of $E_4$ modulates, the variation of EWM is clearly observed. The expressions Eqs. (3) and (8) determine the intensity of the SWM and EWM signals, respectively. The term $\Omega_{\text{SWM}}^1 / \Gamma_{\text{SWM},\nu}$ suppresses the MWM (FWM + SWM + EWM) signals, and $\Omega_{\text{EWM}}^1 / \left[\Gamma_{\text{EWM},\nu \omega} + i(\Delta + \Delta_2)\right]$ plays the dual role of modulating the EWM and suppressing the SWM signals. The field $E_4$ plays the vital role of suppressing and enhancing EWM under the two-photon resonant condition $\Delta + \Delta_2 = 0$. Figure 3(a1) shows the SWM signals, which
unexpectedly decrease with increases of $E_4$ because $\left| \Gamma_{4\omega,4\omega} / \Gamma_{6\omega,6\omega} \right| \ll \left| \Gamma_{4\omega,4\omega} / \left[ \Gamma_{4\omega,4\omega} + i(\Delta_4 + \Delta_1) \right] \right|$ in Eq. (8), which is majorly suppressed by $E_4$ with a smaller $E_1$ power. This result illustrates that under a small power of the probe field, the enhancement effect of $E_4$ is smaller than the suppression effect in Eq. (8).

Fig. 3. MWM signals with different powers of $E_1$, by scanning $\Delta_1$ when $\Delta_1 = -400$ MHz. Signal evolution with fixed $E_1$ power by setting the $E_1$ power to 100 $\mu$W ($\Omega_1 = 2\pi \times 10$ MHz) (a1), 500 $\mu$W ($\Omega_1 = 2\pi \times 54$ MHz) (a2), and 1 mW ($\Omega_1 = 2\pi \times 108$ MHz) (a3). Corresponding dependence curves are shown in (b). Corresponding images of the probe (c) and SWM (d) at different powers of $E_1$ ($\Omega_2 = 2\pi \times 7.6$ MHz; $\Omega_3 = 2\pi \times 142$ MHz; $\Omega_3' = 2\pi \times 224$ MHz; and maximum $\Omega_4 = 2\pi \times 140$ MHz). The color coding is same with Fig. 2.

Figure 3(a2) shows that the EWM signals increase with $P_4$ from 0 mW to 4 mW and the suppression effect of $E_4$ increases; subsequently, the EWM signal disappears when $P_4$ increases from 5 mW to 8 mW. This phenomenon can be easily understood by the competition relation between $\left| \Gamma_{4\omega,4\omega} / \Gamma_{6\omega,6\omega} \right| \ll \left| \Gamma_{4\omega,4\omega} / \left[ \Gamma_{4\omega,4\omega} + i(\Delta_4 + \Delta_1) \right] \right|$ in Eq. (8). Initially, $\left| \Gamma_{4\omega,4\omega} / \Gamma_{6\omega,6\omega} \right| > \left| \Gamma_{4\omega,4\omega} / \left[ \Gamma_{4\omega,4\omega} + i(\Delta_4 + \Delta_1) \right] \right|$, and the enhanced EWM is observed. When the power of $E_4$ is increased to the maximum, the term $\left| \Gamma_{4\omega,4\omega} / \left[ \Gamma_{4\omega,4\omega} + i(\Delta_4 + \Delta_1) \right] \right|$ becomes larger than $\left| \Gamma_{4\omega,4\omega} / \Gamma_{6\omega,6\omega} \right|$, which leads to the suppressed SWM. In Fig. 3(a3), when $P_4$ increases, the suppression dip gradually becomes shallow; then, the EWM peak is observed when $\left| \Gamma_{4\omega,4\omega} / \Gamma_{6\omega,6\omega} \right| \ll \left| \Gamma_{4\omega,4\omega} / \left[ \Gamma_{4\omega,4\omega} + i(\Delta_4 + \Delta_1) \right] \right|$ is satisfied in Eq. (8). This regularity is clearly observed in the dependence curves in Fig. 3(b), where the dots are the experimental results and the solid curves are the theoretical simulations according to Eq. (8). The theoretical subtracting offset is basically consistent with the results of the experiment. Moreover, a chaos-like phenomenon appears in the spectrum signals when $P_4$ is strong as shown in Fig. 3(a). The chaos-like phenomenon in these signals represents the baseline oscillation, and the background of the spectrum varies from peace to noise as shown in the upper part of Fig. 3(a), which is caused directly by the interaction between different orders of MWM and can be attributed to the intrinsic Kerr nonlinearity enhancement in the medium, where dynamic instability and the route to chaos occur [28].

The circularly polarized dressed spatial images are shown in Figs. 3(c) and 3(d). Figure 3(c) shows the probe images with different probe field powers and reflects the intensity of the doubly dressing EIT windows. Figure 3(d) shows the EWM images as the power of the probe
field increases from bottom to top. In these processes, the circular polarization of the probe field is fixed. When the power of the probe field increases from 400 μW to 800 μW, the spatial shift and focusing of the images are observed clearly at approximately Δ_2 = 0 MHz in Fig. 3(d). Based on an analysis of Fig. 2, the polarized nonlinear phase and optical field superposition make the image styles changed. The focusing is attributable to the phase modulation caused by Rydberg interactions. The spatial shift phenomenon, which cannot be observed in the SWM images, is a result of EWM. A comparison of Figs. 3(d3) and 3(d1) shows that the obvious spatial shift is related to EWM. The spatial shift can be described by the nonlinear phase shift equation \[ \phi_2 = 2k_z z n_{2} J_2 e^{(\gamma_{2} - \gamma_{1})t/2} / n_{n} I_{F,S} \]. Based on the equation, the Rydberg interaction leads to a change in \( n_2 \), and FWM and SWM are suppressed; moreover, as \( I_{F,S} \) becomes small, \( \phi_2 \) becomes strong. These results advocate that EWM is more sensitive to Rydberg-Rydberg interaction [19].

![Diagram](image)

Fig. 4. (a) Variation of MWM signals with Δ_4 when Δ_2 is scanned and Δ_1 is fixed at -400 MHz. (b) Theoretical simulation that corresponds to (a). (c) Switch between SWM and EWM by controlling the power of \( E_4 \) by scanning Δ_4. Corresponding Rabi frequency at \( \Omega_1 = 2\pi \times 54 \) MHz; \( \Omega_2 = 2\pi \times 7.6 \) MHz; \( \Omega_3 = 2\pi \times 142 \) MHz; \( \Omega_3' = 2\pi \times 224 \) MHz; and \( \Omega_4 = 2\pi \times 100 \) MHz.

In Fig. 4, we investigate the EWM signals as a function of detuning Δ_4. Figure 4(a) shows the evolution of the Rydberg MWM signal when Δ_2 is scanned at discrete Δ_4. The circularly polarized probe field is appropriately selected so that the Rydberg SWM becomes a pure suppression dip. When |Δ_4| = 100 MHz, a large detuning of \( E_4 \) has a limited effect on the Rydberg SWM because the dressing effect is weaker. When |Δ_4| is tuned to 50 MHz, a spectrum with a half dip and a half peak appears, which illustrates that \( E_4 \) begins to enhance the SWM to generate the observable EWM. When Δ_4 = 0, the two-photon resonant condition is satisfied; therefore, the stronger EWM is observed. Figure 4(a) clearly shows the evolution from the SWM dip to the EWM peak. Figure 4(b) shows the simulated results obtained with the expression of \( \rho^{(7)}_{\nu_4 \nu_4} \) and \( \rho^{(5)}_{\nu_4 \nu_4} \), and the values are scaled according to experimental data. Such trend of simulation results are basically accord with experimental observations. This evolution process is easily controlled in the experiment and can be effectively applied to all-optical switches. Based on this idea as an example of application, we demonstrate different degrees of switching in which the \( E_4 \) field is turned ON and OFF using a manual switch. As shown in Fig. 4(c), with \( E_4 \) ON and OFF, the signals can be switched between SWM dip and EWM peak, and the intensity increases as the power of \( E_1 \) increases.
Finally, to directly control the EWM with polarization, variations of the EWM with the polarization of the dressing field \( E_4 \) are shown in Fig. 5. Figure 5(a1) shows the EWM signals that vary with the polarization of \( E_4 \) in the circularly polarized probe field. In this case, with the scanned \( \Delta_2 \), the dips on the SWM signals are shown at the top right corner of the figure, and each peak represents pure EWM created in the \(|0\rangle \leftrightarrow |1\rangle \leftrightarrow |3\rangle \leftrightarrow |1\rangle \leftrightarrow |2\rangle \leftrightarrow |1\rangle \leftrightarrow |4\rangle \leftrightarrow |1\rangle \) excitation process. When the angle of \( E_4 \) QWP changes from 0° to 45°, the polarization of \( E_4 \) changes from linearly polarized to circularly polarized, which decreases the size of the EWM peaks. This phenomenon can be explained as the dressing effect of the circularly polarized \( E_4 \), which is stronger than the linearly polarized \( E_4 \). The corresponding CG coefficient is calculated and labelled in Fig. 5(c), and \( \left( \Omega_{4u,4}^2 / \Omega_{3u,3}^2 \right)^{1/2} = 3 \) is obtained. As shown in Fig. 5(a1), the height of the EWM signals decreases and subsequently increases because the \( E_4 \) polarization is adjusted from circular into linear when the angle of the QWP changes from 45° to 90°. To avoid the effect of detuning \( \Delta_1 \) on this regularity, Fig. 5(a2) shows the case in Fig. 5(a1) but with \( \Delta_1 = -700 \text{ MHz} \). Compared with Fig. 5(a1), the same regularity is presented, although each EWM peak is smaller because of the larger detuning \( \Delta_1 \). Figure 5(b) shows the dependence curves that correspond to Fig. 5(a1) (black) and Fig. 5(a2) (red), where the dots are experimental observations and solid curves are the theoretic simulations. Here, solid curves are simulated by the overall factors of Eq. (8), and the offset in the experimental observations has been subtracted. The theory is consistent with the experiment.

5. Conclusion

In summary, we directly detect the pure Rydberg EWM and present a series of detailed experiment results on multi-parameter control mechanism. To obtain such Rydberg high order
processes in the experiment is quite challenging. Nonlinearity is small for high-order process, the characters of nonlinear process in our work are technically obtained by electromagnetically induced transparency (EIT) changes with the control fields; EIT enhances the nonlinearities together with polarized filed dressing make high contrast EWM spectrum are extracted directly and then be controlled by the power, detuning, and polarization of the dressing fields. Coexisting SWM and EWM are modulated in different style, in which one is suppression dip, the other is enhancement peak. Such method overcomes the problem that Rydberg EWM always mixes with Rydberg SWM so that relatively strong Rydberg EWM spectrum can be obtained in lab and controlled directly, this controllable EWM process with an ultra-narrow linewidth is sensitive to Rydberg-Rydberg interactions and has the potential to become an effectively non-destructive detection method for investigating the underlying Rydberg property [29] and could be applied in quantum long-distance communication [30]. In addition, the images of the probe and MWM are presented to reflect the nonlinear response of Rydberg–Rydberg interaction. The common cases of focusing and phase shifting are observed, which can reflect Rydberg nonlinear phase variation and establish the relation between refractive index of Rydberg medium and optical field. This study will not only exploit a new method of investigating the phase control of Rydberg nonlinear processes and spatial transmission characteristics, but also provide the basis for the advanced design that is highly dependent on the refractive index materials; more importantly has research value in the field of optical information processing.

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