

Fundamental limits of free-space microwave-to-optical frequency conversion efficiency using Rydberg atoms

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Efficient microwave-to-optical frequency conversion (MOC) using Rydberg atoms is promising for radiometry, electrometry, and hybrid quantum networks. However, in free space, the total system efficiency can be much lower than the internal atomic conversion efficiency because of the large mismatch between microwave wavelength, optical wavelength, and atomic-ensemble size. Here, we develop a systematic theoretical framework in which the total conversion efficiency is factorized into microwave mode-matching efficiency, optical spin-wave extraction efficiency, and internal coherent conversion efficiency. In the regime $\lambda_m \gg L \gg \lambda_o$, the atomic ensemble couples to the microwave field as a collective dipole, whereas its coupling to the optical field is governed by a phase-matched spin-wave mode. We show that the total efficiency is primarily limited by microwave mode matching. For an ideal diffraction-limited Gaussian microwave mode coupled to this collective dipole, the microwave extraction factor is bounded by about $3/16$ under standard polarization and orientation averaging. This analysis provides a transparent benchmark for evaluating free-space Rydberg MOC and identifies microwave mode matching as the key target for experimental optimization.

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I. INTRODUCTION

Microwave and optical frequencies are two complementary wavebands that play crucial roles in modern information technologies [1–3]. On one hand, microwave signals are particularly useful in environment characterization, radar systems, and radiometry for detecting fundamental physics, such as axions and the cosmic microwave background radiation [4,5]. With the advent of quantum information technologies, microwaves have become essential carriers for various quantum systems, including superconducting qubits, spin qubits, and semiconductor double quantum dot qubits [6–8]. Furthermore, employing exotic microwave quantum states has the potential to realize quantum-enhanced microwave technologies with improved sensing and detection sensitivities [9,10]. On the other hand, the optical frequency domain offers advantages such as low noise at room temperature, enabling long-distance communications and efficient single-photon counting [11]. Consequently, quantum transducers that can coherently convert between microwave and optical domains are indispensable for bridging these two important frequency regimes [12,13].

Over the past decade, significant progress has been made in developing integrated photonic waveguide or microresonator systems for coherent microwave-to-optical frequency conversion (MOC). These approaches include electro-optical

interaction [14,15], magneto-optical coupling [16,17], optomechanics [18–20], and doped rare-earth atomic ensembles [21]. These platforms offer huge potential for compact, chip-integrable, and enhanced interactions due to strong light confinement, making them promising for a wide range of applications. In addition to waveguide-based systems, recent experimental demonstrations have shown remarkable advancements in MOC using cold Rydberg atoms [21–27]. Rydberg-atom-based schemes have emerged as promising contenders due to their strong electric dipole coupling and rich electronic structure [28]. Additionally, the Rydberg-atom system also provides unique advantages in realizing free-space MOC, which offers several advantages over waveguide-based approaches. By directly transmitting microwave signals through air, free-space conversion avoids the transmission losses associated with cables and can potentially benefit from the low cosmic background temperature [29,30]. Moreover, Rydberg-atom-based systems can work with a wide angle of free-space microwave modes, providing additional degrees of freedom for detection and communication applications, such as wide-angle receivers and phased arrays for detecting microwave signals [31]. However, the free-space MOC field has been misled by focusing on internal conversion efficiency and celebrating high values (82% in Ref. [27]), while the total system efficiency is significantly limited due to the fundamental geometric mismatch between microwave beams and atomic ensembles.

In this paper, we aim to bridge the gap between the theoretical promise and practical realization of efficient free-space microwave-to-optical conversion using Rydberg

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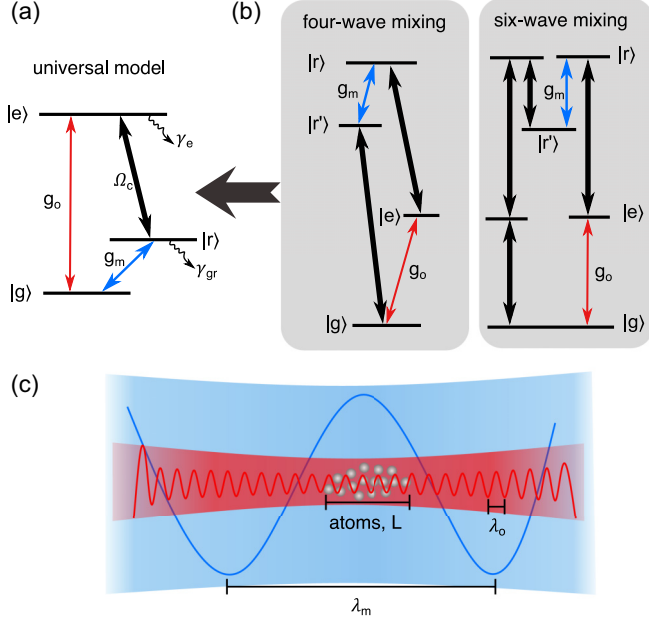


FIG. 1. Schematic of the microwave-to-optical frequency conversion (MOC) based on Rydberg atoms. (a) The Λ -type energy-level diagram as the universal model for MOC with g_m and g_o representing the coupling strengths between the microwave and optical photons, respectively, and an atom. (b) The energy-level diagrams of MOC based on four- and six-wave mixing processes. In (a) and (b), black arrows indicate the coherent drive fields, red arrows denote the optical signal, and blue arrows denote the microwave signal. (c) The illustration of the Rydberg-atom ensemble coupling to the microwave and the optical fields. The blue (red) region and the corresponding solid line indicate the microwave (optical) field, with L , λ_m , $\lambda_o L$, λ_m , and λ_o denoting the ensemble length, microwave wavelength, and optical wavelength, respectively.

atoms. We develop a theoretical framework that factorizes the total system efficiency of free-space Rydberg MOC into microwave-mode extraction, optical-mode extraction, and internal coherent conversion. In the regime $\lambda_m \gg L \gg \lambda_o$, microwave mode matching becomes the dominant bottleneck. For ideal diffraction-limited Gaussian focusing, this yields a microwave extraction bound of about 3/16. Our analysis clarifies the distinction between internal conversion efficiency and total system efficiency, and provides a useful benchmark for evaluating free-space Rydberg MOC.

II. MODEL

The simplified model for coherent MOC in an atomic ensemble is based on a Λ -type energy-level scheme, as illustrated in Fig. 1(a) [22,32]. In this model, a strong coherent drive field (black arrow, Rabi frequency Ω_c) is applied to couple the optical excited state $|e\rangle$ and the intermediate microwave excited state $|r\rangle$. An input optical signal (red arrow) excites the $|g\rangle \leftrightarrow |e\rangle$ transition, and the drive stimulates microwave emission (blue arrow) through the $|r\rangle \leftrightarrow |g\rangle$ transition. Conversely, a microwave input induces optical emission.

While practical implementations employ more complex energy-level structures, such as the four-wave mixing [25,33] and six-wave mixing [24,26,27,34] schemes shown in Fig. 1(b), the fundamental principles remain the same. First, the transitions involved in MOC must form a closed loop, regardless how many energy levels are involved. If the transitions form a chain instead of a loop, a photon conversion will result in a population transfer from the initial state to the end of the transition chain, and induces the entanglement between the converted photon and the excitation in the atomic ensemble, which means decoherence of the converted fields when tracing out the MOC device. Second, the essential mechanism of MOC is nonlinear frequency mixing, where coherent beam-splitter-type interactions between the optical and microwave fields are stimulated by external coherent drives. Specifically, for an m -wave mixing process, a total of $m - 2$ coherent drives are employed to stimulate the MOC process. In waveguide-based MOC schemes, three-wave mixing is realized by electro-optical and magneto-optical effects [14–16], and four-wave mixing is realized in electro-optomechanical systems [35]. This nonlinear susceptibility of dielectrics is usually broadband but weak; the waveguide and resonator structures are utilized to enhance the interactions. In contrast, the atomic-ensemble system exploits near-resonant transitions to achieve strong nonlinear optical susceptibility for multi-wave mixing. Based on the two principles, all atom-based MOC schemes can be reduced to the effective model shown in Fig. 1(a). Through adiabatic elimination of intermediate energy levels under large detuning conditions (see Appendix A for a detailed derivation), the four-wave and six-wave mixing schemes shown in Fig. 1(b) can be reduced to an effective Λ -type system. In this reduced picture, the microwave coupling between $|r\rangle$ and $|r'\rangle$ in the original scheme is transformed into an effective coupling between $|g\rangle$ and $|r\rangle$, with renormalized coupling strengths that incorporate the effects of the eliminated intermediate states.

Figure 1(c) illustrates the cold-atom ensemble mediating the coherent coupling between the free-space microwave and optical beams. This system is particularly interesting due to the difference in the wavelengths of microwave ($\lambda_m \sim \text{cm}$) and optical ($\lambda_o \sim \mu\text{m}$) photons compared to the size of the atomic ensemble ($L \sim \text{mm}$), with the relationship $\lambda_m \gg L \gg \lambda_o$. This disparity in scales leads to distinct collective light-matter interaction behavior, prompting a separate investigation of the microwave beam-atom and optical beam-atom couplings in the following.

III. MICROWAVE COUPLING

As depicted in Fig. 1(a), the atoms couple to the microwave field through the levels $|g\rangle$ and $|r\rangle$. Since $L \ll \lambda_m$, atoms can be approximated as a point and the collective behavior of atoms could be treated as a single harmonic oscillator with a weak input microwave field, referred to as a “superatom” in Fig. 2(a). The excitation in an N -atom ensemble can be represented by the bosonic operator $S^\dagger = \frac{1}{\sqrt{N}} \sum_j \sigma_j^+$ according to the Holstein-Primakoff approximation [36], where subscript j denotes the j th atom and $\sigma^+ = |r\rangle\langle g|$ is the raising operator. Here, the approximation applies when $N \gg 1$ and the excitation number is small, and also the Rydberg blockade

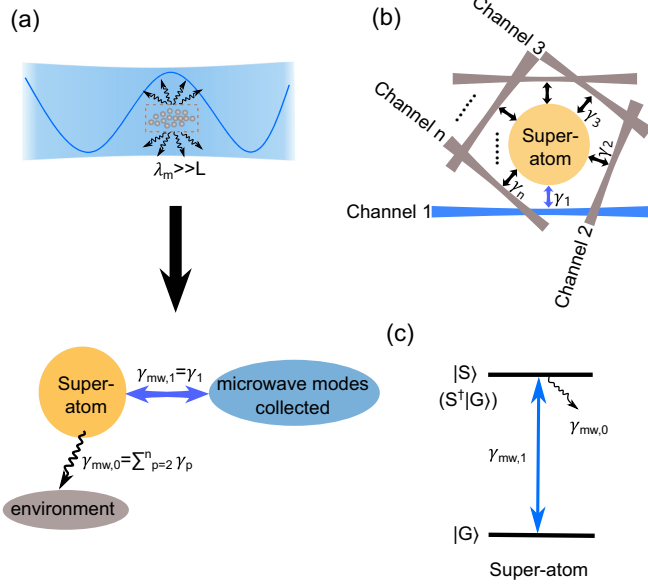


FIG. 2. (a) The atomic-ensemble coupling with microwaves can be treated as a superatom due to $\lambda_m \gg L$. According to the multichannel model shown in (b), the coupling can be simplified as in the bottom of panel (a). The superatom emits microwave into the target modes with rate $\gamma_{mw,1}$ and into the undesired modes with rate $\gamma_{mw,0}$. The ratio $\gamma_{mw,1}/\gamma_{mw,0}$ is determined by the focusing of the target microwave. (b) The multichannel model illustrates the coupling of free-space continuum modes with the superatom. The free-space microwave field modes coupled to the superatom can be divided into two parts: one can be collected (blue) and the other cannot (black). (c) Energy levels of the single excited superatom. The superatom and microwave are coupled with a rate of $\gamma_{mw,1}$.

effect between atoms is negligible due to the large mean distance between excited atoms compared to the blockade radius [37,38]. The ground state of the system is denoted as $|G\rangle = |gg \cdots gg\rangle = |g^{\otimes N}\rangle$, and the single excitation state is given by $|R\rangle = S^\dagger |G\rangle$, with a collective amplitude decay rate due to spontaneous emission (SE) of $\gamma_{mw} = N\gamma_{gr}$ [39,40], where γ_{gr} is the SE rate of a single-atom transition from $|r\rangle$ to $|g\rangle$.

The conversion process in Rydberg atoms can be described by the scattering between different radiation channels of the superatom, as described by Fig. 2(b). Each channel represents a continuum of modes, such as a Gaussian beam in free space. The conversion efficiency is determined by the ratio of the SE rate into a target continuum mode to that of the undesired modes [41]. For example, as shown in Fig. 2(b), the emission rate into the target continuous mode (blue in channel 1) is γ_1 and that into the undesired modes (gray) is $\sum_{p=2}^n \gamma_p$; thus, the conversion efficiency is determined by $\gamma_1 / \sum_{p=1}^n \gamma_p$. To obtain a specific expression for the radiation rate γ_1 , the coupling system between superatom and multiple channels is solved. The multichannel coupling shown on the right of Fig. 2(a) is described by the Hamiltonian

$$H_m = \omega_s S^\dagger S + \sum_{p=1}^n \sum_k [\omega_{p,k} b_{p,k}^\dagger b_{p,k} + g_{p,k} b_{p,k}^\dagger S + g_{p,k}^* b_{p,k} S^\dagger],$$

where ω_s is the effective transition frequency of the superatom, and $\omega_{p,k}$ and $b_{p,k}$ are the frequency and bosonic operator of the microwave mode with wave vector k in the p th channel, respectively, given the coupling strength $g_{p,k}$ with the superatom. Applying the Markovian and Born approximations, the steady-state solution (see Appendix A for details) for the superatom operator under external fields $b_{p,in}$ with frequency ω is given by

$$S(\omega) = \frac{-i \sum_{p=1}^n \sqrt{2\gamma_p} b_{p,in}}{-i(\omega - \omega_s) + \gamma_R + \sum_{p=1}^n \gamma_p}, \quad (1)$$

where $\gamma_p = \pi \rho_{p,k} g_{p,k}^2$ (details shown in Appendix A) represents the SE rate of superatoms into the p th channel, $\rho_{p,k}$ is the mode density in channel p , and γ_R is the amplitude decay rate due to nonradiative decay (such as atomic collisions, and irregular motion of atoms inside superatoms).

Based on the above, the coupling between atoms and microwaves is presented as the bottom of Fig. 2(a), where the superatom emits the microwave into the target modes with the rate $\gamma_{mw,1}$ and into the undesired modes with rate $\gamma_{mw,0}$. Given the target microwaves focused on the atoms is a Gaussian beam with a beam waist w_0 , the emission rate into the continuous Gaussian beam is $\gamma_{mw,1} = (3\lambda_m^2/4\pi^2 w_0^2) \gamma_{mw}$ [see Eqs. (B16) and (B21)], where $\gamma_{mw} = N\gamma_{gr}$ is the sum of the emission rates of superatom into all microwave modes. As mentioned before, the conversion efficiency will be determined by the ratio $\gamma_{mw,1}/\gamma_{mw}$. Thus, the focusing of the microwave (characterized by the waist w_0) can limit the conversion efficiency. Finally, the coupling between the superatom and target microwave is shown in Fig. 2(c), where the coupling rate is $\gamma_{mw,1}$ and the decay rate of the superatom is $\gamma_{mw,0}$.

IV. OPTICAL COUPLING

Since the ensemble size is much larger than the optical wavelength, $L \gg \lambda_o$, the collective effect of the optical coupling differs from the microwave coupling, as shown in Fig. 3(a). To describe the collective interactions, we introduce the concept of a collective spin-wave mode, defined as $E_{\vec{k}} = \frac{1}{\sqrt{N}} \sum_j e^{i\vec{k} \cdot \vec{r}_j} \sigma_{ge,j}$, where $\sigma_{ab} = |a\rangle \langle b|$ ($a, b \in [g, r, e]$) is the localized atomic jump operator, $\sigma_{ab,j}$ is the corresponding operator at position $\vec{r}_j$, and $\vec{k}$ is the wave vector. The spin-wave mode is essentially a Fourier transformation of localized atomic excitations. Consequently, the coupling between atoms and the optical modes reads

$$H_o = \frac{|g_o|}{\sqrt{N}} \sum_{\vec{k}_0} \sum_{\vec{k}_1} \sum_{j=1}^N e^{i(\vec{k}_0 - \vec{k}_1) \cdot \vec{r}_j} e^{i(\omega_{a,\vec{k}_0} - \omega_{ge})t} a_{\vec{k}_0}^\dagger E_{\vec{k}_1} + \text{H.c.}, \quad (2)$$

where g_o is the coupling strength between a single atom and the plane-wave optical modes, $a_{\vec{k}_0}$ denotes the annihilation operators of optical photons, and $\vec{k}_{0,1}$ denotes the wave vector of the optical (spin-wave) mode. This Hamiltonian shows that there are multiple collective atomic excitation couples with various optical modes, making the general treatment of the optical coupling with the atomic ensemble complex. Instead of using a multiple-channel model as studied with the microwave

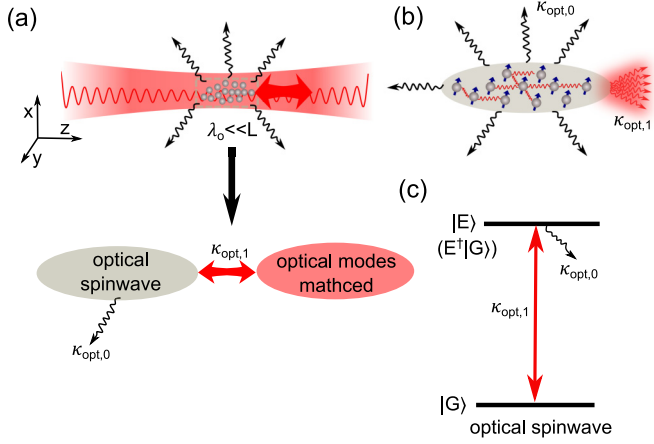


FIG. 3. (a) The coupling between the atomic ensemble and optical fields with $\lambda_0 \ll L$ can be simplified as the coupling between the optical spin wave and the matched free-space optical radiation beam, according to the collective effect shown in (b). (b) Schematic of the principle of directional superradiance. Atoms form a phase-ordered collective excited state of spin waves through coherent coupling. Influenced by the cigar-shaped spatial distribution of the ensemble, the collective decay of spin-wave excitation leads to directional emission into the matched free-space optical beam, with a spontaneous emission rate of $\kappa_{\text{opt},1}$. The optical spin wave also emits photons into undesired modes with rate $\kappa_{\text{opt},0}$. When the optical depth of the atomic ensemble is much greater than 1, $\kappa_{\text{opt},1} \gg \kappa_{\text{opt},0}$. (c) Energy levels of singly excited optical spin wave, which couples with the optical field at rate $\kappa_{\text{opt},1}$.

coupling case [Fig. 2(b)], we introduce a free-space optical radiation beam that matches the target spin-wave mode, as illustrated on the right of Fig. 3(a). The essential difference for the coupling model originates from the distinct scale of the wavelength and the atomic ensemble, where the constructive interference between individual atoms leads to directional optical radiation, as shown in Fig. 3(b), which has been studied in Refs. [42,43]. In an atomic ensemble where interatomic spacing is comparable to the radiation wavelength, atoms establish coherent coupling through their shared radiation field. This process allows the phase information of incident light to be collectively stored as an optical spin wave. Upon collective deexcitation, the atoms radiate photons with the same phase. As shown in Fig. 3(b), for a cigar-shaped ensemble, this results in constructive interference exclusively along the major axis, producing a highly directional superradiant beam whose spatial mode is determined by the spatial distribution of atoms. The free-space radiation beam has a size significantly larger than the wavelength, and the optical setup could be efficiently modulated to perfectly match the profile of the radiation mode. Then, by approximating the spatial density function of the atomic ensemble as a Gaussian distribution, we can reconstruct the one-dimensional free-space beam mode that is emitted by the spin-wave excitation, with its bosonic operator, as $r_{k_0}^\dagger = \frac{1}{A} \sum_{k_{0,z}} \sum_{\theta} e^{-\frac{1}{2}(k_0^2 - k_{0,z}^2)\sigma^2} a_{k_0}^\dagger$. Here, A is a normalization factor, and σ is the width of the atomic cloud in the optical light propagation (z) direction (more details are provided in Appendix C). For this radiation mode, we can derive the coupling between the spin-wave mode with

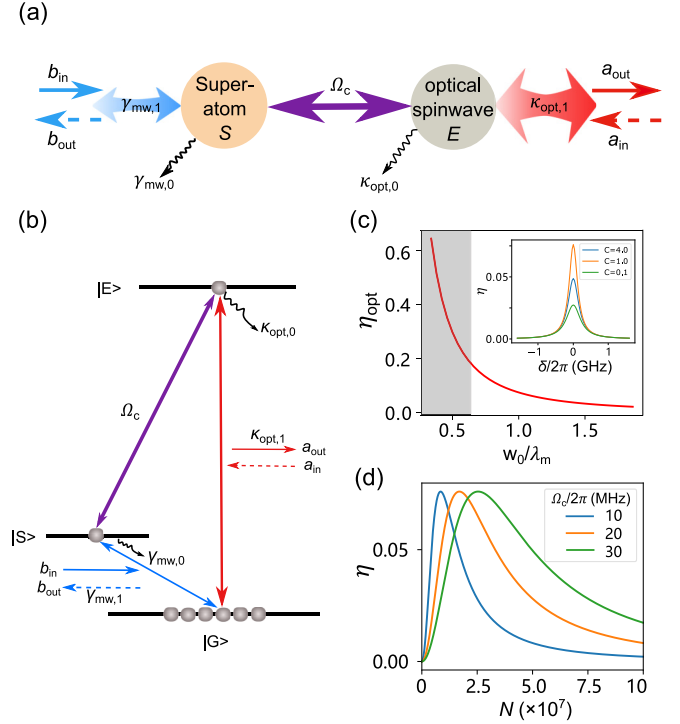


FIG. 4. (a) Schematic of the conversion between microwave and optical signals by coupling the superatom and spin-wave modes. (b) Energy-level diagram for MOC. (c) The fundamental limit on optimal conversion efficiency (η_{opt}) imposed by the microwave beam waist (w_0). Inset: The dependence of η on the frequency detuning δ for different cooperativities (C). (d) Conversion efficiency (η) as a function of atom number N for various Rabi frequencies Ω_c .

the continuum as the SE rate $\kappa_{\text{opt},1} = \frac{d_{\text{opt}}}{4} \gamma_e$, where γ_e is the SE rate of a single atom, and d_{opt} is the mean optical depth (OD) of the atomic ensemble in the z direction. In this work, we consider $d_{\text{opt}} \gg 1$ [42,43] (see Appendix C). Compared with the multiple-channel model for microwave coupling, optical coupling can be described by a single-channel model. However, other effects, such as the diffusion of atoms that changes the locations and collisions between atoms, can induce an extra decay rate $\kappa_{\text{opt},0}$ to the spin wave. Thus, the optical spin wave emits photons into the target modes at rate $\kappa_{\text{opt},1}$ and into the undesired modes at rate $\kappa_{\text{opt},0}$, as shown in Fig. 3(a). Moreover, the directional emission rate $\kappa_{\text{opt},1}$ due to the collective effect is much larger than the decay rate $\kappa_{\text{opt},0}$ when $d_{\text{opt}} \gg 1$. The optical spin wave couples with the matched optical field at a rate of $\kappa_{\text{opt},1}$, as shown as Fig. 3(c), where the decay rate of the optical spin wave is $\kappa_{\text{opt},0}$.

V. CONVERSION EFFICIENCY

As illustrated in Fig. 4(a), by combining the input-output models for the atomic ensemble's coupling to microwave and optical free-space beams, we can realize MOC by introducing a coherent coupling between the superatom and spin-wave excitations. Through the control light field with Rabi frequency

Ω_c , the MOC system can be described by the Hamiltonian

$$H = \delta E^\dagger E + \Omega_c (S^\dagger E + S E^\dagger) + [\sqrt{2\gamma_{mw,1}} S^\dagger b_{in} + \sqrt{2\kappa_{opt,1}} E^\dagger a_{in} + \text{H.c.}], \quad (3)$$

where $\delta = \omega_e - \omega_a$ is the detuning between the optical transition (ω_e) and the optical signal frequency ω_a . The corresponding energy-level diagram is shown in Fig. 4(b), where $|G\rangle$ is the ground state, $|R\rangle$ is the superatom singly excited state, and $|E\rangle$ is the singly excited state of the optical spin wave. We assume the microwave signal is on resonance with the microwave transition and the external drive matches the frequency difference between optical and microwave signals. The details about the derivation of the above conversion Hamiltonian are provided in Appendix D.

The MOC model enables bidirectional frequency conversion between microwave and optical photons. For either input, a part of the excitation can be converted to the desired electromagnetic wave frequency band with a quantum efficiency given by

$$\eta = \frac{\gamma_{mw,1}}{\gamma_{mw} + \gamma_R} \frac{\kappa_{opt,1}}{\kappa_{opt,0} + \kappa_{opt,1}} \frac{4C}{(1+C)^2 + \delta^2/\kappa_{opt}^2}, \quad (4)$$

where $C = \Omega_c^2/[(\gamma_R + \gamma_{mw})(\kappa_{opt,0} + \kappa_{opt,1})]$ is the cooperativity for the coherent coupling between the superatom and the spin-wave modes. It is worth noting that this expression resembles the results for other cavity-enhanced MOC [12]. The first two terms represent extraction factors, determined by the ratio between the external coupling rate and the total decay rate of the modes, corresponding to the impedance matching or mode matching in practical experiments. The third term is the internal conversion efficiency, reaching unity when $C = 1$ and $\delta = 0$. Therefore, the maximum achievable η is intrinsically limited by the extraction factors. Notably, the distinct coupling models for microwave (multichannel) and optical (single-channel) fields reveal a crucial asymmetry: while the optical extraction factor can approach unity due to directional superradiance when $d_{opt} \gg 1$, the microwave extraction factor is fundamentally limited by diffraction-constrained focusing. This asymmetry, arising from the different length scales $\lambda_m \gg L \gg \lambda_o$, identifies microwave mode matching as the fundamental bottleneck for free-space MOC efficiency.

Considering that the internal decay rate $\kappa_{opt,0}$ can be much smaller than the emission rate of the optical spin wave, i.e., $\kappa_{opt,0} \ll \kappa_{opt,1}$, in a cold-atom ensemble by optimizing its temperature and density, and the ratio between the Rydberg state's intrinsic decay and stimulated decay (γ_R/γ_{mw}) can be optimized to approach zero via increasing the coherent drive strengths, the eventual η is limited by the extraction ratio of the microwave port as [for details see Eqs. (B16) and (B21) and their associated discussion]

$$\eta \leq \frac{\gamma_{mw,1}}{\gamma_{mw}} = \frac{3\lambda_m^2}{4\pi^2 w_0^2}. \quad (5)$$

Physically, the microwave extraction factor $\gamma_{mw,1}/\gamma_{mw}$ is determined by the focusing capability of the collection optics. As shown in Eq. (B21), $\gamma_{mw,1} \propto 1/A_{\text{eff}}$, where A_{eff} is the effective focal area of the microwave beam. A lens with higher numerical aperture (NA) produces a smaller A_{eff} , thereby

increasing the extraction factor. However, A_{eff} is fundamentally bounded by the diffraction limit: $A_{\text{eff}} \geq \pi(2\lambda_m/\pi)^2 \simeq \pi(0.61\lambda_m)^2$ for $\text{NA} \rightarrow 1$. This gives $\gamma_{mw,1}/\gamma_{mw} \leq 3/16$ for ideal diffraction-limited Gaussian focusing. In the regime $\lambda_m \gg L$, where the atomic ensemble couples to the microwave field as a collective dipole, this factor sets the maximum total efficiency when the internal conversion and optical extraction efficiencies approach unity. Figure 4(c) shows the achievable optimal η_{opt} for a given microwave beam waist w_0 in free space, with the shaded area denoting the region forbidden by the diffraction limit. While experiments like Ref. [27] achieve impressive internal conversion efficiency of 82% without optical cavities, the total system efficiency can be estimated as $\eta = 82\% \times S_M/S_{mw} \simeq 2.2 \times 10^{-4}$ [27], which remains fundamentally limited by microwave mode matching to 3/16, regardless of atomic performance.

The bound in Eq. (5) applies when the ensemble acts as a single collective microwave dipole, i.e., for $\lambda_m \gg L$. A natural way to enhance the microwave extraction factor is to increase the longitudinal size of the ensemble and enter the extended-ensemble regime. In this case, the phase variation of the microwave field along the atomic cloud must be taken into account, and the simple single-superatom description is no longer sufficient. In Appendix E, we provide an approximate treatment by dividing a long atomic cloud into a chain of short slices, each of which is described as a local superatom, and by cascading their transfer matrices. This slicing analysis shows that, when the longitudinal length becomes comparable to λ_m , the conversion efficiency can exceed the 3/16 bound derived for a single collective microwave dipole and reach about 25% for parameters close to those of Ref. [27]. This does not contradict Eq. (5), because Eq. (5) applies to the regime $\lambda_m \gg L$, where the ensemble responds as a single collective dipole coupled to a far-field Gaussian microwave mode. We also note that increasing the transverse size is not a straightforward extension within the present model. The optical extraction factor used above relies on directional emission from a phase-matched optical spin wave in an elongated, cigar-shaped ensemble, as discussed around Fig. 3 and derived in Appendix C. A large transverse size would modify the spatial profile of the optical radiation mode and can reduce the coupling efficiency from the spin-wave excitation to the target optical mode assumed in the single-channel optical model. Moreover, if the transverse diameter becomes comparable to λ_m , the microwave phase and polarization vary across all spatial dimensions. A fully three-dimensional treatment of microwave phase, polarization, radiation-mode matching, and optical spin-wave emission is then required, and the corresponding efficiency bound is expected to be geometry dependent. Besides increasing the ensemble size, microwave cavities can enhance the microwave extraction factor, although at the cost of reduced free-space bandwidth and angular acceptance. Microwave near-field engineering may provide another possible route, but its quantitative performance requires a dedicated model including antenna geometry, impedance matching, and dissipative losses.

To evaluate the MOC performance under different conditions, we numerically simulated a ^{87}Rb atomic ensemble [27]. The microwave beam, with a beam waist $w_0 = \lambda_m$, is

coupled to the Rydberg transition $|40P_{3/2}\rangle \rightarrow |39D_{5/2}\rangle$, while the optical beam is coupled to the transition $|5S_{1/2}\rangle \rightarrow |5P_{3/2}\rangle$. According to the fundamental limit [Eq. (5)], we can only achieve an efficiency of 7.6%. The inset of Fig. 4(c) shows the conversion efficiency against the detuning for various C , showing the conversion bandwidth of about 0.32, 0.26, and 0.45 GHz for $C = 4, 1$, and 0.1 , respectively, which is mainly determined by $\kappa_{\text{opt},1}$ with $\Omega_c/2\pi = 20$ MHz. For a fixed atomic transition and $\delta = 0$, C linearly increases with $\Omega_c^2 N$, with the corresponding OD of the atomic ensemble estimated with a transverse atomic cloud waist of 66 μm . Figure 4(d) shows that η reaches its maximal value for an optimal N . A larger N means a larger $C > 1$, inhibiting MOC due to coupling-induced coherent oscillations when exchanging the excitation between superatom and spin wave, equivalent to a spectral splitting in the response making the system's optimal working condition deviating from $\delta = 0$. Similarly, C is also determined by Ω_c^2 , and the optimal N increases with increasing Ω_c . The parameters in Fig. 4 assume negligible Rydberg blockade, which is justified both by the low excitation regime of quantum applications and by the flexibility to adjust atomic density and Rabi frequency to avoid blockade effects.

VI. CONCLUSION

We have developed a theoretical framework for evaluating the total system efficiency of free-space Rydberg MOC. In this framework, the efficiency is factorized into microwave-mode extraction, optical-mode extraction, and internal coherent conversion efficiency. In the regime $\lambda_m \gg L \gg \lambda_o$, the framework shows that microwave mode matching is the dominant bottleneck and leads to a maximum microwave extraction factor of about 3/16 for ideal diffraction-limited Gaussian focusing. This result provides a well-defined benchmark for the regime $\lambda_m \gg L$, where the atomic ensemble couples to the microwave field as a single collective dipole. When the ensemble length becomes comparable to λ_m , longitudinal phase variation of the microwave field becomes important, and a geometry-dependent extended-ensemble treatment is required. Our work suggests that near-field microwave engineering remains an interesting future direction in realizing high-efficiency quantum interfaces based on free-space atomic ensembles, which have the potential to enable a wide range of applications in quantum technologies, including quantum communication, quantum sensing, and hybrid quantum systems [8,44,45]. But its impact on the total conversion efficiency requires a dedicated quantitative analysis. Furthermore, the theoretical methods developed here can be extended to other applications involving disparate length scales of electromagnetic modes, such as the fundamental limitation of the detection sensitivity of electrometry [9,10], and imaging and spectroscopy in the terahertz regime [46,47].

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DATA AVAILABILITY

The data that support the findings of this article are available from the corresponding author upon reasonable request.

APPENDIX A: SIMPLIFIED MODEL FOR MULTIWAVE MIXING

This section explains the simplified models for four-wave and six-wave mixing processes, as depicted in Fig. 1(b) of the main text.

1. Four-wave mixing

Shown in Fig. 5 is the schematic of the simplified Raman transition for microwave input, which stimulates the transition directly from $|g\rangle$ to $|r\rangle$. For the system of four-wave mixing, the Hamiltonian is given by

$$H = \Delta|r'\rangle\langle r'| + (\Omega|g\rangle\langle r'| + \Omega_m|r'\rangle\langle r| + \text{H.c.}). \quad (\text{A1})$$

Here, $\Delta \gg \Omega, \gamma_r$. For large detuning, we have $\rho_{r'r'} \approx 0$, and $\rho_{gr'}$ and $\rho_{r'r}$ are at their steady states:

$$\begin{aligned} \rho_{gr'} &\simeq \frac{\Omega}{-\Delta} \rho_{gg} + \frac{\Omega_m}{-\Delta} \rho_{gr}, \\ \rho_{r'r} &\simeq \frac{\Omega}{-\Delta} \rho_{gr} + \frac{\Omega_m}{-\Delta} \rho_{rr}. \end{aligned} \quad (\text{A2})$$

Substituting the steady results of $\rho_{gr'}$ and $\rho_{r'r}$ into the differential equation for ρ_{gr} , we can obtain

$$\frac{d\rho_{gr}}{dt} = -i\Omega\rho_{r'r} + i\Omega_m\rho_{gr'} - \frac{\gamma_r}{2}\rho_{gr}$$

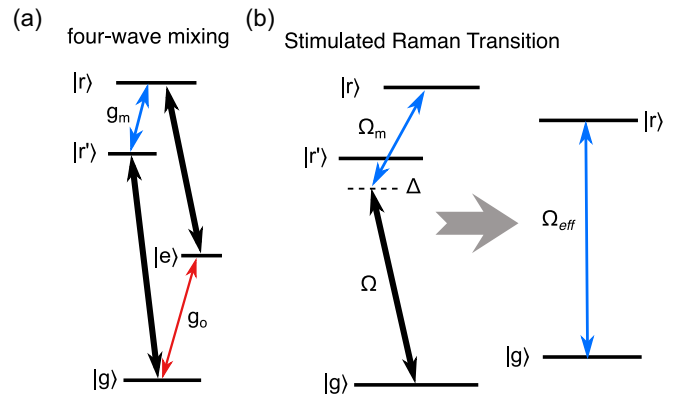


FIG. 5. The schematic of the stimulated Raman transitions for the microwave input in a four-wave mixing model.

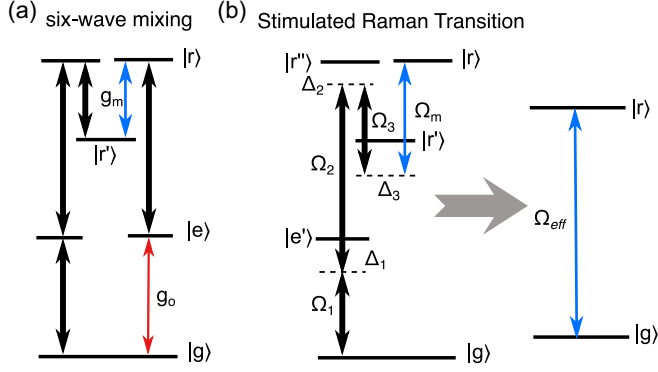


FIG. 6. The schematic of the stimulated Raman transitions for the microwave input in a six-wave mixing model.

$$\simeq -i\Omega_{\text{eff}}(\rho_{gg} - \rho_{rr}) + \left[i \left(\frac{\Omega^2}{\Delta} - \frac{\Omega_m^2}{\Delta} \right) - \frac{\gamma}{2} \right] \rho_{gr}, \quad (\text{A3})$$

where $\Omega_{\text{eff}} = \Omega\Omega_m/\Delta$ is the effective Rabi frequency for the four-wave mixing process.

2. Six-wave mixing

In Fig. 6, the model for the six-wave mixing system is illustrated. The Hamiltonian is given by

$$H = \Delta_1|e'\rangle\langle e'| + \Delta_2|r''\rangle\langle r''| + \Delta_3|r'\rangle\langle r'| + (\Omega_1|g\rangle\langle e'| + \Omega_2|e'\rangle\langle r''| + \Omega_3|r'\rangle\langle r''| + \Omega_m|r'\rangle\langle r| + \text{H.c.}). \quad (\text{A4})$$

Here $\Delta_1, \Delta_2, \Delta_3 \gg \Omega_1, \Omega_2, \Omega_3, \Omega_m, \gamma_{e'}, \gamma_{r''}, \gamma_{r'}$. Thus, we can assume that $\rho_{ii} \approx 0$ for intermediate state $i \in [e', r'', r']$, and ρ_{ij} ($i, j \in [g, e', r'', r', r]$, $ij \neq gg, rr, gr, rg$) are at their steady states. Substituting these steady results into the differential equation for ρ_{gr} , we can obtain

$$\frac{d\rho_{gr}}{dt} \simeq -i\Omega_{\text{eff}}(\rho_{gg} - \rho_{rr}), \quad (\text{A5})$$

where $\Omega_{\text{eff}} = \Omega_1\Omega_2\Omega_3\Omega_m/(\Delta_1\Delta_2\Delta_3)$ is the effective Rabi frequency for the six-wave mixing process.

These simplified models provide valuable insights into the dynamics of four-wave and six-wave mixing processes in multilevel atomic systems.

APPENDIX B: MICROWAVE COUPLING

1. Superatom coupled to a single microwave channel

The excitation of the superatom, S , as well as the input-output theory for the free-space microwave coupling with the superatom, requires the understanding of its coupling to a continuum. Such a continuum could be the three-dimensional vacuum modes in free space or the free-space propagation Gaussian beam mode. We first consider the simplest model, with the superatom coupled to a one-dimensional continuum, i.e., a single microwave channel, as

$$H = \omega_s S^\dagger S + \sum_k [\omega_k b_k^\dagger b_k + g_k b_k^\dagger S + g_k^* b_k S^\dagger]. \quad (\text{B1})$$

Here, the continuum of microwave modes is described by the variable k , which corresponds to the wave vector of a

wave; b_k describes the bosonic annihilation operator of the continuum mode, ω_k is its frequency that is determined by the dispersion relationship of the continuum, and g_k is the coupling strength between the superatom and the continuum mode. The coupling strength could be solved by applying the box quantization of the continuum mode, and the g_k is $\sqrt{N}$ times as large as that of the single atom due to the collective enhancement effect. Consequently, the evolution of operators S and b_k follow

$$\frac{db_k}{dt} = i[H, b_k] = -i\omega_k b_k - ig_k S, \quad (\text{B2})$$

$$\frac{dS}{dt} = i[H, S] \simeq -i\omega_s S - i \sum_k g_k b_k, \quad (\text{B3})$$

where the Holstein-Primakoff approximation $[S, S^\dagger] \simeq 1$ is applied for the superatom excitation S . Introducing the transformation

$$b_k = e^{-i\omega_k t} f_k(t), \quad (\text{B4})$$

we obtain

$$\frac{df_k(t)}{dt} = -ig_k e^{i\omega t} S. \quad (\text{B5})$$

Then, we can get the formal solution of $f(t)$ as

$$f_k(t) = f_k(t_0) + \int_{t_0}^t [-ig_k e^{i\omega_k t'} S(t')] dt', \quad (\text{B6})$$

and obtain the solution of b_k as

$$b_k = e^{-i\omega_k(t-t_0)} b_{k,in} - ig_k \int_{t_0}^t e^{i\omega_k(t'-t)} S(t') dt', \quad (\text{B7})$$

where $b_{k,in} = e^{-i\omega_k t_0} f_k(t_0)$. Substituting the solution of b_k into the evolution equation of S , we can obtain

$$\begin{aligned} \frac{dS}{dt} = & -i\omega_s S - i \int_0^\infty \rho_k g_k \left[e^{-i\omega_k(t-t_0)} b_{k,in} \right. \\ & \left. - ig_k \int_{t_0}^t e^{i\omega_k(t'-t)} S(t') dt' \right] dk, \end{aligned} \quad (\text{B8})$$

where the sum of microwave modes is replaced by the integral $\sum_{k=0}^\infty \rightarrow \int_0^\infty \rho_k dk$, with ρ_k denoting the mode density of a one-dimensional microwave field. Since only one direction of the propagating microwave field is considered, the integral of k ranges from zero to infinity.

Introducing the input field operator

$$b'_{in}(t) = \frac{1}{2\pi} \int_{-\infty}^\infty e^{-i\omega(t-t_0)} b_{\omega,in} d\omega, \quad (\text{B9})$$

the dynamics of S follow

$$\begin{aligned} \frac{dS}{dt} = & -i\omega_s S - ig_\omega \rho_\omega 2\pi b'_{in} \\ & - \int_{-\infty}^\infty \rho_\omega g_\omega^2 \int_{t_0}^t e^{-i\omega(t-t')} S(t') dt' d\omega. \end{aligned} \quad (\text{B10})$$

By exchanging the order of integration of time and frequency and using the relations

$$\int_{-\infty}^\infty e^{-i\omega(t-t')} d\omega = 2\pi \delta(t-t'),$$

$$\int_{t_0}^t f(t')\delta(t-t')dt' = \frac{1}{2}f(t), \quad (\text{B11})$$

we arrive at

$$\frac{dS}{dt} = (-i\omega_s - \pi\rho_\omega g_\omega^2)S - i2\pi\rho_\omega g_\omega b'_{\text{in}}. \quad (\text{B12})$$

Eventually, by defining the external coupling rate between the superatom and channel as $\gamma_{\text{mw},1} = \pi\rho_\omega g_\omega^2$, and the corresponding input fields from the channel $b_{\text{in}} = \sqrt{2\pi\rho_\omega}b'_{\text{in}}$, we obtain the input-output relation in a standard form as

$$\frac{dS}{dt} = (-i\omega_s - \gamma_{\text{mw},1})S - i\sqrt{2\gamma_{\text{mw},1}}b_{\text{in}}, \quad (\text{B13})$$

with the transmission

$$b_{\text{out}} = b_{\text{in}} - i\sqrt{2\gamma_{\text{mw},1}}S. \quad (\text{B14})$$

At steady state, the superatom excitation under a microwave signal input at frequency ω is

$$S(\omega) = \frac{i\sqrt{2\gamma_{\text{mw},1}}b_{\text{in}}(\omega)}{-i(\omega_s - \omega) - \gamma_{\text{mw},1}}. \quad (\text{B15})$$

For one-dimensional continuous modes with the effective mode area at the cross section as A_{eff} , we have $\rho_\omega = \frac{1}{2\pi c}$ and $\hbar g_\omega = \sqrt{N}d_{gr}\sqrt{\frac{\hbar\omega}{2\epsilon_0 A_{\text{eff}}}}$; then we have the coupling rate

$$\gamma_{\text{mw},1} = \pi\rho_\omega g_\omega^2 = \frac{Nd_{gr}^2\omega}{4c\hbar\epsilon_0 A_{\text{eff}}}. \quad (\text{B16})$$

Here d_{gr} is the electric dipole moment of the atomic transition $|g\rangle \rightarrow |r\rangle$. Here A_{eff} corresponds to the focal area of the lens for the target microwave mode, $A_{\text{eff}} = \pi w_0^2$, where w_0 is the waist radius of the focused Gaussian beam. Under the lens's diffraction-limited conditions, the minimum focal area attains a value of $A_{\text{eff}} = \pi(2\lambda_{\text{mw}}/\pi)^2$, approximately equivalent to the Airy disk area of $\pi(0.61\lambda_{\text{mw}})^2$ at NA = 1, thereby establishing an upper bound for $\gamma_{\text{mw},1}$.

2. Superatom coupled to multiple channels

The free-space microwave modes can be viewed as the sum of multiple-channel microwave fields. Thus, the coupling between the superatom and the microwave fields in free space can be described as

$$H = \omega_s S^\dagger S + \sum_{p=1}^n \sum_k [\omega_{p,k} b_{p,k}^\dagger b_{p,k} + g_{p,k} b_{p,k}^\dagger S + g_{p,k}^* b_{p,k} S^\dagger]. \quad (\text{B17})$$

Here the subscript p denotes the p th channel. The evolution equations of operators are

$$\begin{aligned} \frac{db_{p,k}}{dt} &= i[H, b_{p,k}] = -i\omega_{p,k}b_{p,k} - ig_{p,k}S, \\ \frac{dS}{dt} &= i[H, S] \simeq -i\omega_s S - i \sum_{p=1}^n \sum_k g_{p,k} b_{p,k}. \end{aligned} \quad (\text{B18})$$

Taking the same method as for single-channel coupling to obtain the solution of S , we can obtain

$$\begin{aligned} \frac{dS}{dt} &= \left(-i\omega_s - \sum_{p=1}^n [\pi\rho_{p,\omega} g_{p,\omega}^2] \right) S - i \sum_{p=1}^n [2\pi\rho_{p,\omega} g_{p,\omega} a_{p,\text{in}}] \\ &= \left(-i\omega_s - \sum_{p=1}^n \gamma_p \right) S - i \sum_{p=1}^n \sqrt{2\gamma_p} b_{p,\text{in}}. \end{aligned} \quad (\text{B19})$$

Through Fourier transform, the steady solution of S is

$$S = \frac{-i \sum_{p=1}^n \sqrt{2\gamma_p} b_{p,\text{in}}(\omega)}{-i(\omega - \omega_s) + \sum_{p=1}^n \gamma_p}. \quad (\text{B20})$$

We can find that the form of this static solution of S is consistent with that of the absorption spectra of a single atom. The coupling rate $\sum_{p=1}^n \gamma_p$ between the superatom and the microwave modes in the total channel, noted as γ_R , is N times the rate of spontaneous radiation of a single atom γ_{gr} in free space, i.e.,

$$\gamma_R = \sum_{p=1}^n \gamma_p = N\gamma_{gr} = N \frac{d_{gr}^2 \omega_{gr}^3}{6\pi \hbar \epsilon_0 c^3}. \quad (\text{B21})$$

Here d_{gr} and ω_{gr} are the electric dipole moment and frequency, respectively, of the atomic transition $|g\rangle \rightarrow |r\rangle$.

APPENDIX C: OPTICAL COUPLING

For the coupling between optical fields and the atomic ensemble, we only consider a simplified two-level model, with transition frequency ω_{ge} and spontaneous emission rate γ_e .

1. The spin-wave mode

The interaction between optical fields and atoms, considering all free-space plane-wave optical modes, can be described by the Hamiltonian

$$H = \sum_{\vec{k}_0} \sum_{j=1}^N g_{j,\vec{k}_0} a_{\vec{k}_0}^\dagger \sigma_{ge,j} e^{i(\omega_{a,\vec{k}_0} - \omega_{ge})t} + \text{H.c.}, \quad (\text{C1})$$

where $\vec{k}_0$ is the vector of plane waves and $\sigma_{ge,j}$ is the jump operator for the j th atom, and the summation is over all optical modes and atoms at different locations. Introducing the spin-wave mode of atomic excitation with very low excitation numbers, i.e.,

$$E_{\vec{k}} = \frac{1}{\sqrt{N}} \sum_j e^{i\vec{k}\cdot\vec{r}_j} \sigma_{ge,j}, \quad (\text{C2})$$

$$\sigma_{ge,j} = \frac{1}{\sqrt{N}} \sum_{\vec{k}} e^{-i\vec{k}\cdot\vec{r}_j} E_{\vec{k}}, \quad (\text{C3})$$

the Hamiltonian can be rewritten in momentum space as

$$H = \frac{|g|}{\sqrt{N}} \sum_{\vec{k}_0} \sum_{\vec{k}_1} \sum_{j=1}^N e^{i(\vec{k}_0 - \vec{k}_1)\cdot\vec{r}_j} e^{i(\omega_{a,\vec{k}_0} - \omega_{ge})t} a_{\vec{k}_0}^\dagger E_{\vec{k}_1} + \text{H.c.} \quad (\text{C4})$$

For a cold atomic ensemble with a spatial distribution approximated by Gaussian distributions, we can replace the

summation with the integration of the density function:

$$\begin{aligned} \sum_j^N &= N \iiint \rho_{\text{density}} dx dy dz \\ &= N \iiint \frac{1}{\sqrt{2\pi}^3} e^{-\frac{x^2}{2\sigma_x^2}} e^{-\frac{y^2}{2\sigma_y^2}} e^{-\frac{z^2}{2\sigma_z^2}} dx dy dz. \end{aligned} \quad (\text{C5})$$

The space integral yields

$$\int_{-\infty}^{\infty} \frac{1}{\sqrt{2\pi}\sigma_x} e^{-\frac{x^2}{2\sigma_x^2}} e^{i(k_{0,x}-k_{1,x})x} dx \simeq e^{-\frac{1}{2}(k_{0,x}-k_{1,x})^2\sigma_x^2}. \quad (\text{C6})$$

For cold atomic systems with a cigar-shaped spatial distribution, $\sigma_x, \sigma_y \ll \sigma_z$. Thus, for a determined probe light a_{k_0} , any spin wave in the x and y directions can be effectively coupled to that signal light, while only optical spin waves consistent with the signal light, i.e., $k_{1,z} \simeq k_{0,z}$, can be efficiently excited. It is because σ_x and σ_y are small enough to satisfy $(k_{0,x}-k_{1,x})^2\sigma_x^2 \simeq 0$ and $(k_{0,y}-k_{1,y})^2\sigma_y^2 \simeq 0$, and $\sigma_z \gg \sigma_x, \sigma_y$.

We consider only the optical spin-wave mode with vector $\vec{k}_z$, and assume that the spin-wave modes are not coupled to each other, i.e., each spin-wave mode is considered to be independently coupled to the light-field modes, while the moving and dephasing effects of atoms are neglected. Then, the coupling Hamiltonian in momentum space can be simplified as

$$H = \sqrt{N}|g| \sum_{\vec{k}_0} \sum_{\vec{k}_1} a_{\vec{k}_0}^\dagger E_{\vec{k}_1} e^{-\frac{1}{2}[(\vec{k}_0-\vec{k}_1)\vec{\sigma}]^2} e^{i(\omega_{a,\vec{k}_0}-\omega_{ge})t} + \text{H.c.} \quad (\text{C7})$$

In the frame of polar coordinates, we can rewrite

$$\sum_{\vec{k}_0} \rightarrow \sum_{k_0} \sum_{k_{0,z}} \sum_{\theta}, \quad (\text{C8})$$

where $k_{0,x}$ and $k_{0,y}$ can be presented as

$$k_{0,x} = \sqrt{k_0^2 - k_{0,z}^2} \cos \theta, \quad k_{0,y} = \sqrt{k_0^2 - k_{0,z}^2} \sin \theta. \quad (\text{C9})$$

For the optical spin wave with wave vector $\vec{k}_z$, the coupling Hamiltonian between the optical spin wave $E_{\vec{k}_1}$ and the optical fields can be shown as

$$\begin{aligned} H &= \sqrt{N}|g| \sum_{\vec{k}_0} [a_{\vec{k}_0}^\dagger E_{\vec{k}_1} e^{-\frac{1}{2}[(\vec{k}_0-\vec{k}_1)\vec{\sigma}]^2} e^{i(\omega_{a,\vec{k}_0}-\omega_{ge})t} + \text{H.c.}] \\ &= \sqrt{N}|g| \sum_{\theta} \sum_{k_0} \sum_{k_{0,z}} [e^{-\frac{1}{2}k_{0,x}^2\sigma^2} e^{-\frac{1}{2}k_{0,y}^2\sigma^2} e^{-\frac{1}{2}(k_{0,z}-k_{1,z})^2\sigma_z^2} \\ &\quad \times e^{i(k_0-k_{ge})ct} k_0 a_{\vec{k}_0}^\dagger E_{\vec{k}_1} + \text{H.c.}] \\ &\simeq A\sqrt{N}|g| \sum_{k_0=-\infty}^{\infty} [e^{-\frac{1}{2}(k_0-k_1)^2\sigma_z^2} e^{i(k_0-k_{ge})ct} k_0 r_{k_0}^\dagger E_{\vec{k}_1} + \text{H.c.}], \end{aligned} \quad (\text{C10})$$

where

$$r_{k_0}^\dagger = \frac{1}{A} \sum_{k_{0,z}} \sum_{\theta} e^{-\frac{1}{2}(k_0^2-k_{0,z}^2)\sigma^2} a_{\vec{k}_0}^\dagger \quad (\text{C11})$$

is a reconstructed bosonic operator of the optical field, whose form is related to the spatial distribution ρ_{density} of the atomic system and A is the normalization factor. According to the commutation relation $[r_{k_0}^\dagger, r_{k_0}] = -1$, we can get

$$\begin{aligned} [r_{k_0}^\dagger, r_{k_0}] &= \frac{1}{|A|^2} \sum_{k_{0,z}} \sum_{\theta} e^{-(k_0^2-k_{0,z}^2)\sigma^2} [a_{\vec{k}_0}^\dagger, a_{\vec{k}_0}] \\ &= \frac{1}{|A|^2} \int_0^{k_0} \int_0^{2\pi} \rho e^{-(k_0^2-(k_0-\delta)^2)\sigma^2} d\delta d\theta \\ &\simeq \frac{1}{|A|^2} \int_0^{k_0} \int_0^{2\pi} \rho e^{-(k_0^2-k_0^2+2k_0\delta)\sigma^2} d\delta d\theta \\ &= \frac{1}{|A|^2} 2\pi \rho_{2D} \frac{1 - e^{-2k_0^2\sigma^2}}{2k_0\sigma^2} \\ &\simeq \frac{1}{|A|^2} \frac{\pi \rho_{2D}}{k_0\sigma^2} \\ &= -1. \end{aligned} \quad (\text{C12})$$

Thus, we derive the normalization factor as

$$|A|^2 \simeq \frac{\pi \rho_{2D}}{k_0\sigma^2}, \quad (\text{C13})$$

where ρ_{2D} is the optical mode density of two-dimensional free space, $\delta = k_0 - k_{0,z}$, and $\sigma_x = \sigma_y = \sigma$.

2. The evolution of the system

Based on the effective Hamiltonian derived in the previous section, the evolution of the operators can be described as

$$\begin{aligned} \frac{dE_{\vec{k}_1}}{dt} &= -iA\sqrt{N}|g| \sum_{k_0} e^{-\frac{1}{2}(k_0-k_1)^2\sigma_z^2} k_0 r_{k_0}, \\ \frac{dr_{k_0}}{dt} &= -i(k_0 - k_{ge})cr_{k_0} - iA\sqrt{N}|g| e^{-\frac{1}{2}(k_0-k_1)^2\sigma_z^2} k_0 E_{\vec{k}_1}. \end{aligned} \quad (\text{C14})$$

According to the differential equation for r_{k_0} , the solution should be of the form

$$r_{k_0} = e^{-i(k_0-k_{ge})ct} f(t). \quad (\text{C15})$$

Comparing the differentials of this form with the evolution equation for r_{k_0} as

$$\begin{aligned} \frac{dr_{k_0}}{dt} &= -i(k_0 - k_{ge})cr_{k_0} + e^{-i(k_0-k_{ge})ct} \frac{df(t)}{dt}, \\ \frac{dr_{k_0}}{dt} &= -i(k_0 - k_{ge})cr_{k_0} - iA\sqrt{N}|g| e^{-\frac{1}{2}(k_0-k_1)^2\sigma_z^2} k_0 E_{\vec{k}_1}, \end{aligned} \quad (\text{C16})$$

we obtain

$$\frac{df(t)}{dt} = -ie^{i(k_0-k_{ge})ct} A\sqrt{N}|g| e^{-\frac{1}{2}(k_0-k_1)^2\sigma_z^2} k_0 E_{\vec{k}_1}. \quad (\text{C17})$$

Integrating this expression from the initial moment t_0 gives

$$\begin{aligned} f(t) &= f(t_0) - i \int_{t_0}^t e^{i(k_0-k_{ge})ct'} A\sqrt{N}|g| \\ &\quad \times e^{-\frac{1}{2}(k_0-k_1)^2\sigma_z^2} k_0 E_{\vec{k}_1}(t') dt'. \end{aligned} \quad (\text{C18})$$

Substituting this expression back into the solution for r_{k_0} , we obtain

$$r_{k_0} = e^{-i(k_0 - k_{ge})c(t-t_0)} r_{k_0, \text{in}} - iA\sqrt{N}|g| \times e^{-\frac{1}{2}(k_0 - k_1)^2 \sigma_z^2} \int_{t_0}^t e^{-i(k_0 - k_{ge})c(t-t')} k_0 E_{\vec{k}_1}(t') dt', \quad (\text{C19})$$

where $r_{k_0, \text{in}} = e^{-i(k_0 - k_{ge})ct_0} f(t_0)$ denotes the optical field input at the initial moment t_0 . Substituting the solution into the evolution equation of the optical spin-wave operator, we can obtain

$$\begin{aligned} \frac{dE_{\vec{k}_1}}{dt} = & -iA\sqrt{N}|g| \int_{-\infty}^{\infty} \rho_{1D} e^{-\frac{1}{2}(k_0 - k_1)^2 \sigma_z^2} e^{-i(k_0 - k_{ge})c(t-t_0)} \\ & \times k_0 r_{k_0, \text{in}} dk_0 - A^2 N |g|^2 \int_{-\infty}^{\infty} \rho_{1D} e^{-(k_0 - k_1)^2 \sigma_z^2} \\ & \times \int_{t_0}^t e^{-i(k_0 - k_{ge})c(t-t')} k_0 E_{\vec{k}_1}(t') dt' dk_0, \end{aligned} \quad (\text{C20})$$

where the summation over k_0 has been replaced by the integral

$$\sum_{k_0=-\infty}^{\infty} \rightarrow \int_{-\infty}^{\infty} \rho_{1D} dk_0, \quad (\text{C21})$$

with ρ_{1D} being the optical mode density of one-dimensional free space. The mode density of the one-dimensional free space, ρ_{1D} , is related to the mode density of the two-dimensional free space, ρ_{2D} , by

$$\rho_{1D} \rho_{2D} = 2 \frac{L_x L_y L_z}{(2\pi)^3 c}, \quad (\text{C22})$$

where the factor of 2 accounts for the two polarizations, and $L_{x,y,z}$ are the box quantization lengths in the x , y , and z directions, respectively.

Similar to the treatment based on the Weisskopf-Wigner approximation [48], the coupling terms are insensitive to k_0 for $k_0 \approx k_{ge}$. We can therefore replace k_0 with k_{ge} in Eq. (C20), simplifying the integral over k_0 as

$$\int_{-\infty}^{\infty} e^{-i(k_0 - k_{ge})c(t-t')} dk_0 = 2\pi \delta(t - t'). \quad (\text{C23})$$

Applying the following equality of the integral over t ,

$$\int_{t_0}^t f(t') \delta(t - t') dt' = \int_{t_0}^t f(t') \delta(t - t') dt' = \frac{1}{2} f(t), \quad (\text{C24})$$

we obtain the following evolution equation:

$$\frac{dE_{\vec{k}_1}}{dt} = -i\sqrt{N}gA_{\text{in}}(t) - \kappa_{\text{opt},1} E_{\vec{k}_1}(t). \quad (\text{C25})$$

Here, $A_{\text{in}}(t)$ is a reconstructed annihilation operator of the input optical field, whose form is related to the spatial distribution ρ_{density} of the atomic system, denoted as

$$A_{\text{in}}(t) = A \int_{-\infty}^{\infty} \rho_{1D, \omega} e^{-\frac{1}{2}(k_0 - k_1)^2 \sigma_z^2} e^{-i(k_0 - k_{ge})c(t-t_0)} k_0 r_{k_0, \text{in}} dk_0, \quad (\text{C26})$$

and

$$\begin{aligned} \kappa_{\text{opt},1} &= e^{-(k_{ge} - k_1)^2 \sigma_z^2} \pi \rho_{1D} N g_{\omega}^2 |A|^2 k_{ge} \\ &= e^{-(k_{ge} - k_1)^2 \sigma_z^2} \pi 2 \frac{L_x L_y L_z}{8\pi^3 c} N d_{ge}^2 \frac{\omega}{4\hbar \epsilon_0 L_x L_y L_z} \frac{\pi}{\sigma^2} \\ &\simeq e^{-(k_{ge} - k_1)^2 \sigma_z^2} \frac{N d_{ge}^2 \omega_{ge}}{16\pi \hbar \epsilon_0 c \sigma^2} \end{aligned} \quad (\text{C27})$$

represents the decay rate of $E_{\vec{k}_1}$.

3. The mean optical depth d_{opt}

The atomic number is directly related to the optical depth of the atomic system. The mean optical depth d_{opt} of the atomic system in the direction of optical light propagation is calculated as [49]

$$\begin{aligned} d_{\text{opt}} &= \frac{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{\frac{-x^2}{2\sigma_x^2}} e^{\frac{-y^2}{2\sigma_y^2}} dx dy \int_{-\infty}^{\infty} N \rho_{\text{density}} dz \sigma_{\text{scr}}}{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{\frac{-x^2}{2\sigma_x^2}} e^{\frac{-y^2}{2\sigma_y^2}} dx dy} \\ &= \frac{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{\frac{-x^2}{2\sigma_x^2}} e^{\frac{-y^2}{2\sigma_y^2}} dx dy \int_{-\infty}^{\infty} \frac{N}{\sqrt{2\pi^3} \sigma_x \sigma_y \sigma_z} e^{\frac{-z^2}{2\sigma_z^2}} dz \sigma_{\text{scr}}}{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} e^{\frac{-x^2}{2\sigma_x^2}} e^{\frac{-y^2}{2\sigma_y^2}} dx dy} \\ &= \frac{\pi \sigma_x \sigma_y \frac{N}{\sqrt{2\pi^3} \sigma_x \sigma_y} \sigma_{\text{scr}}}{2\pi \sigma_x \sigma_y} = \frac{N \sigma_{\text{scr}}}{4\pi \sigma_x \sigma_y}, \end{aligned} \quad (\text{C28})$$

where $\sigma_{\text{scr}} = 3\lambda_{ge}^2/(2\pi)$ is the optical absorption cross section. According to this equation, we can express the atomic number in the ensemble as

$$N = \frac{4\pi \sigma_x \sigma_y d_{\text{opt}}}{\sigma_{\text{scr}}} = \frac{4\pi \sigma^2 d_{\text{opt}}}{\sigma_{\text{scr}}}. \quad (\text{C29})$$

Substituting this expression into Eq. (C27), which describes the decay rate of the optical spin-wave mode, we obtain

$$\frac{\kappa_{\text{opt},1}}{\gamma_e} = \frac{3}{8\sigma^2} \frac{4\pi \sigma^2 d_{\text{opt}}}{\sigma_{\text{scr}}} \left(\frac{\lambda_{ge}}{2\pi} \right)^2 = \frac{d_{\text{opt}}}{4}, \quad (\text{C30})$$

where $\gamma_e = d_{ge}^2 \omega_{ge}^3 / 6\pi \hbar \epsilon_0 c^3$ is the amplitude decay rate of the excited state $|r\rangle$ of a single atom.

APPENDIX D: EFFECTIVE CONVERSION HAMILTONIAN

The original Hamiltonian is

$$\begin{aligned} H = & \sum_{j, k_a, k_b} \omega_s \sigma_{rr}^j + \omega_e \sigma_{ee}^j + \omega_a a_k^\dagger a_k + \omega_b b_k^\dagger b_k \\ & + (g_a \sigma_{ge}^j a_k^\dagger + g_b \sigma_{gr}^j b_k^\dagger + \Omega_c \sigma_{re}^j e^{i\omega_c t} + \text{H.c.}). \end{aligned} \quad (\text{D1})$$

With the rotating frame of $H_0 = \omega_a a_k^\dagger a_k + \omega_b b_k^\dagger b_k + \omega_a \sigma_{ee} + \omega_b \sigma_{rr}$, the Hamiltonian can be shown as

$$H = \sum_{j, k_a, k_b} \delta \sigma_{ee}^j + (g_a \sigma_{ge}^j a_k^\dagger + g_b \sigma_{gr}^j b_k^\dagger + \Omega_c \sigma_{re}^j + \text{H.c.}), \quad (\text{D2})$$

where $\sigma_{ab} = |a\rangle\langle b|$ ($a, b \in [g, r, e]$) is the localized atomic jump operator and $\sigma_{ab,j}$ is the corresponding operator for the j th atom. Here, we take $\omega_b = \omega_s$, $\omega_a = \omega_b + \omega_c$, and $\delta =$

$\omega_e - \omega_a$. The above Hamiltonian consists of four parts: the eigenenergy $\sum_j \sigma_{ee}^j$, the optical coupling $\sum_{j,k_a} g_a \sigma_{ge}^j a_k^\dagger$, the microwave coupling $\sum_{j,k_b} g_b \sigma_{gr}^j b_k^\dagger$, and the driving coupling $\sum_j \Omega_c \sigma_{re}^j$. Using the operators of the superatom transition ($S^\dagger, S$) and the optical spin wave ($E^\dagger, E$), the Hamiltonian can be represented.

With the relation between atomic transition operators and optical spin-wave operators, we can get

$$\begin{aligned} \sum_j \delta \sigma_{ee}^j &= \sum_j \delta \sigma_{eg}^j \sigma_{ge}^j \\ &= \sum_j \delta \frac{1}{\sqrt{N}} \sum_{\vec{k}_1} e^{i\vec{k}_1 \vec{r}_j} E_{\vec{k}_1}^\dagger \frac{1}{\sqrt{N}} \sum_{\vec{k}_2} e^{-i\vec{k}_2 \vec{r}_j} E_{\vec{k}_2} \\ &= \delta \int \rho_{\text{density}} d\vec{r} \sum_{\vec{k}_1, \vec{k}_2} e^{i(\vec{k}_1 - \vec{k}_2) \vec{r}_j} E_{\vec{k}_1}^\dagger E_{\vec{k}_2} \\ &\simeq \delta \sum_k E_k^\dagger E_k. \end{aligned} \quad (\text{D3})$$

Here, considering the fact that the size of the atomic space distribution is much larger than the optical wavelength, we can assume that $\vec{k}_1 \simeq \vec{k}_2$.

The optical coupling can be represented by replacing atomic operators with optical spin-wave operators, which is the same as Eq. (2) in the main text. The coupling optical modes a can be divided into two parts for analysis, one of which can be collected and excited while the other cannot. The coupling of uncollectible modes with optical spin wave leads to spin-wave loss, as Eq. (C25). Thus, the coupling of all optical modes and optical spin waves in free space can be expressed in a simplified way as

$$\begin{aligned} \sum_{j, \vec{k}_a} g_a \sigma_{ge}^j a_k^\dagger &= \frac{|g|}{\sqrt{N}} \sum_{\vec{k}_a} \sum_{\vec{k}_1} \sum_{j=1}^N e^{i(\vec{k}_a - \vec{k}_1) \vec{r}_j} a_{\vec{k}_a}^\dagger E_{\vec{k}_1} \\ &\simeq \sum_{k_1} g_{a, \text{eff}} a_{\text{in}}^\dagger E_{k_1} - i\kappa_{\text{opt},1} E_{k_1}^\dagger E_{k_1}. \end{aligned} \quad (\text{D4})$$

Here the coupling rate of the input optical modes satisfies $g_{a, \text{eff}} \leq \sqrt{2\kappa_{\text{opt},1}}$. To obtain the optimal conversion efficiency, the ideal $g_{a, \text{eff}} \simeq \sqrt{2\kappa_{\text{opt},1}}$ is adopted.

For the microwave coupling, with the definition of $|G\rangle = |g^{\otimes N}\rangle$, $|R\rangle = \frac{1}{\sqrt{N}} \sum_j |gg \cdots r_j \cdots gg\rangle$, $S = |G\rangle\langle R|$, and Eq. (B19), we can obtain

$$\begin{aligned} \sum_{j, k_b} g_b \sigma_{gr}^j b_k^\dagger &= \sum_k \sqrt{N} g_b |G\rangle\langle R| b_k^\dagger \\ &= \sum_{p=1}^n \sqrt{2\gamma_{pw,1}} b_{p, \text{in}}^\dagger S - i \sum_{p=1}^n \gamma_p S^\dagger S. \end{aligned} \quad (\text{D5})$$

When the input microwave field is modeled as one-dimensional continuum modes, we can obtain

$$\sum_{j, k_b} g_b \sigma_{gr}^j b_k^\dagger = \sqrt{2\gamma_{mw,1}} b_{\text{in}}^\dagger S - i \sum_{p=1}^n \gamma_p S^\dagger S. \quad (\text{D6})$$

Considering the case where only one atom can be excited to an excited state and the Rabi frequency Ω_c of the control

field is much larger than the decay rate γ_e of the excited state $|e\rangle$ ($\Omega_c \gg \gamma_e$) [50,51], with the definition of $S^\dagger = |R\rangle\langle G|$ and the relation between σ_{eg} and optical spin wave E ($\sigma_{ge, j} = \frac{1}{\sqrt{N}} \sum_{\vec{k}} e^{-i\vec{k} \cdot \vec{r}_j} E_{\vec{k}}$), we can obtain

$$\begin{aligned} \sum_j \Omega_c e^{ik_c r_j} \sigma_{re}^j &= \sum_j \Omega_c e^{ik_c r_j} \sigma_{rg}^j \sigma_{ge}^j \\ &= \sqrt{N} \Omega_c S^\dagger \frac{1}{\sqrt{N}} \sum_k e^{-i(k-k_c) r_j} E_k \\ &\simeq \sum_k \Omega_c S^\dagger E_k, \end{aligned} \quad (\text{D7})$$

with approximation $|G\rangle\langle G| = I$. Here we also take the approximation that $e^{-i(k-k_c) r_j} \simeq 1$ for small atomic size and small difference between k_c and k .

Given that the optical spin waves $\sum_k E_k$ are independently coupled to the optical fields, the original Hamiltonian [Eq. (D2)] can be rewritten as

$$\begin{aligned} H &= \delta E^\dagger E + (\sqrt{2\kappa_{\text{opt},1}} a_{\text{in}}^\dagger E + \sqrt{2\gamma_{mw,1}} b_{\text{in}}^\dagger S \\ &\quad + \Omega_c S^\dagger E + \text{H.c.}), \end{aligned} \quad (\text{D8})$$

which is the same as Eq. (3) in the main text.

APPENDIX E: METHOD OF SLICING

In the main text, we only consider a model with length of the atomic cloud much smaller than the microwave wavelength. However, in practical experiments, there is the potential to realize a much longer atomic cloud by weakening the confinement of a magneto-optical trap in the z direction [27,52]. Here, we show that such a situation can also be approximately solved with our model by the method of “slicing,” i.e., treating the long atomic cloud as a chain of many short slices, where each slice could be treated as a single superatom. Therefore, the MOC of the entire atomic cloud can be obtained by solving cascaded slices. For each slice, the MOC can be represented by a transfer matrix $\mathbf{T}$ for the forward-propagating optical and microwave modes, i.e.,

$$\begin{pmatrix} a_{\text{out}} \\ b_{\text{out}} \end{pmatrix} = \mathbf{T} \begin{pmatrix} a_{\text{in}} \\ b_{\text{in}} \end{pmatrix}. \quad (\text{E1})$$

According to the input-output relationships

$$\begin{aligned} a_{\text{out}} &= a_{\text{in}} - i\sqrt{2\kappa_{\text{opt},1}} E, \\ b_{\text{out}} &= b_{\text{in}} - i\sqrt{2\gamma_{mw,1}} S, \end{aligned} \quad (\text{E2})$$

we obtain the elements of the transfer matrix as

$$\mathbf{T} = \begin{pmatrix} \frac{i\delta/\kappa_{\text{opt},1} + C - 1}{i\delta/\kappa_{\text{opt},1} + C + 1} & \frac{i2\Omega_c}{\gamma_R} \sqrt{\frac{\gamma_{mw,1}}{\kappa_{\text{opt},1}}} \\ \frac{i2\Omega_c}{\gamma_R} \sqrt{\frac{\gamma_{mw,1}}{\kappa_{\text{opt},1}}} & -i2 \frac{\delta\gamma_{mw,1}}{\kappa_{\text{opt},1}\gamma_R} - 2 \frac{\gamma_{mw,1}}{\gamma_R} + i \frac{\delta}{\kappa_{\text{opt},1}} + C + 1 \end{pmatrix}. \quad (\text{E3})$$

When the atomic ensemble is divided into n slices, the final output should be obtained as

$$\begin{pmatrix} a_{\text{out}} \\ b_{\text{out}} \end{pmatrix} = \mathbf{T}^n \begin{pmatrix} a_{\text{in}} \\ b_{\text{in}} \end{pmatrix}. \quad (\text{E4})$$

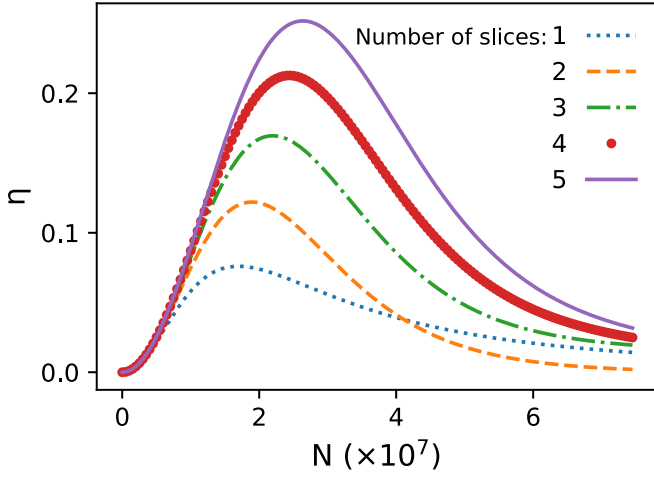


FIG. 7. The dependence of conversion efficiency η on the total atom number N for various atomic-ensemble slices, with each slice having a length of $\lambda_m/2$; the parameters are the same as in the main text.

Thus, the conversion efficiency from microwave to optical frequency is $\eta = |\mathbf{T}^n[0, 1]|^2$.

Taking the same parameters as Fig. 4 in the main text, the conversion efficiency for different numbers of slices is calculated and the result is shown in Fig. 7. Here, we define the length of each slice as $\lambda_m/2$, within which the microwave transition of atoms approximately has a uniform coupling strength and phase across all free-space microwave modes, and a total of N atoms are equally divided across all slices. The results indicate that when the number of atoms is smaller than 7×10^6 , the conversion efficiency η is insensitive to the length of the atomic cloud, $L = n\lambda_m/2$. However, when the atom number is larger, η can be improved by increasing the length of the atomic ensemble while η for a single superatom is limited due to $C > 1$. We find that the dependence of η on N for different slice number n shows similar behavior, as there is an optimal N . For the experimental situation in Ref. [27], where the length of the atomic ensemble is $L = 21$ mm, the highest achieved η can be improved to 25% as $n \approx 5$ for $\lambda_m = 8$ mm.

APPENDIX F: THE LIMITS OF FREQUENCY CONVERSION EFFICIENCY FOR DIFFERENT SITUATIONS

To better understand that the fundamental limit of free-space microwave-to-optical frequency conversion efficiency is due to the fact that the radiation rate of the “superatom” is uniform in all directions, we compare the upper-bound conversion efficiency of “optical-to-optical” (“microwave-to-microwave”) to that of “microwave-to-optical.”

1. Optical-to-optical conversion

When both wavelengths of optical fields (incident and converted optical fields) are much smaller than the atomic-ensemble size, the medium for frequency conversion is no longer ‘superatomic-optical spin wave’ [as shown in Fig. 4(a) of the main text] but “optical spin wave–optical spin wave”

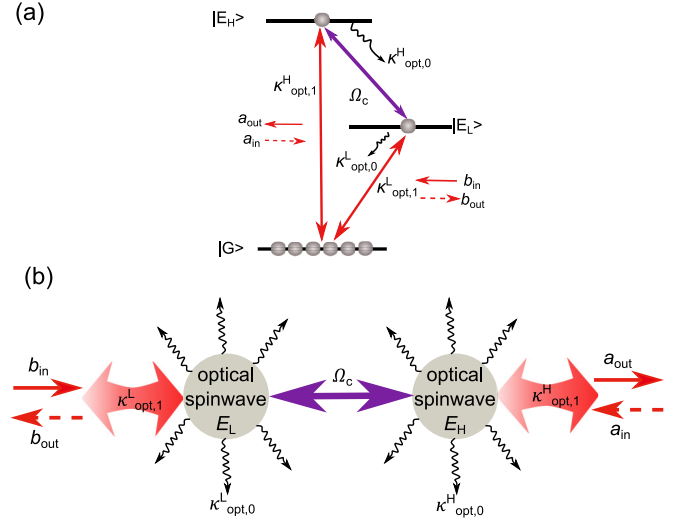


FIG. 8. (a) Energy-level diagram and (b) schematic of the conversion from optical to optical. Here, both optical field modes a and b have the frequency of the optical wave. They couple with the atomic ensemble with rate $\kappa_{\text{opt},1}^H$ and $\kappa_{\text{opt},1}^L$ to form optical spin-wave excitations, denoted as $|E_H\rangle$ and $|E_L\rangle$, respectively.

(as shown in Fig. 8). Similar to the introduction and treatment of “optical coupling” in the main text, we can use the collective spin-wave mode $E_L = \frac{1}{\sqrt{N}} \sum_j e^{i\vec{k}_{gr} \cdot \vec{r}_j} \sigma_{gr,j}$ ($E_H = \frac{1}{\sqrt{N}} \sum_j e^{i\vec{k}_{ge} \cdot \vec{r}_j} \sigma_{ge,j}$) to describe the collective interactions between atoms and the corresponding optical field due to $\lambda_{gr} \ll L$ ($\lambda_{ge} \ll L$). The ground state of both optical spin waves is $|G\rangle$ and the excited states are $|E_L\rangle = E_L^\dagger |G\rangle$ and $|E_H\rangle = E_H^\dagger |G\rangle$, respectively, as shown in Fig. 8(a). Two optical spin waves exhibit enhanced radiation in the direction of atomic extension with radiation rates $\kappa_{\text{opt},1}^L = \frac{d_{\text{opt}}^L}{4} \gamma_r$ and $\kappa_{\text{opt},1}^H = \frac{d_{\text{opt}}^H}{4} \gamma_e$, respectively, and radiation in other directions with radiation rates $\kappa_{\text{opt},0}^L$ and $\kappa_{\text{opt},0}^H$, respectively. $d_{\text{opt}}^{L,H}$ are the mean optical depths of the atomic ensemble in the z direction for optical fields with wavelengths λ_L and λ_H , respectively. When $d_{\text{opt}}^{L(H)} \gg 1$, $\kappa_{\text{opt},0}^{L(H)}$ are much smaller than $\kappa_{\text{opt},1}^{L(H)}$.

Similar to the conversion schematic of MOC introduced in Sec. V of the main text, we can realize optical-to-optical conversion (OOC) by introducing a coherent coupling between the two optical spin waves; the corresponding schematic is shown in Fig. 8(b). By a control light field with Rabi frequency Ω_c , the OOC system can be described by the Hamiltonian

$$H = \delta E_H^\dagger E_H + \Omega_c (E_L^\dagger E_H + E_L E_H^\dagger) + [\sqrt{2\kappa_{\text{opt},1}^L} E_L^\dagger b_{\text{in}} + \sqrt{2\kappa_{\text{opt},1}^H} E_H^\dagger a_{\text{in}} + \text{H.c.}], \quad (\text{F1})$$

where $\delta = \omega_e - \omega_a$ is the detuning between the optical transition (ω_e) and the optical signal frequency ω_a while another optical field (ω_b) is on resonance with the corresponding transition (ω_r) and the external drive matches the frequency difference between the two optical fields.

The OOC model enables bidirectional frequency conversion between two optical frequencies and the quantum

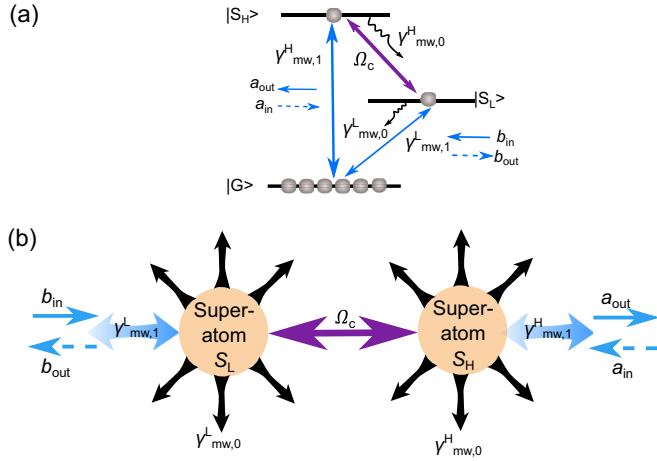


FIG. 9. (a) Energy-level diagram and (b) schematic of the conversion from microwave to microwave. Here, both microwave modes a and b have the frequency of the microwave. They couple with the atomic ensemble with rate $\gamma_{mw,1}^H$ and $\gamma_{mw,1}^L$, inducing the excitations of the corresponding superatom, denoted as $|S_H\rangle$ and $|S_L\rangle$, respectively.

conversion efficiency is

$$\eta_{OOC} = \frac{\kappa_{opt,1}^L}{\kappa_{opt,0}^L + \kappa_{opt,1}^L} \frac{\kappa_{opt,1}^H}{\kappa_{opt,0}^H + \kappa_{opt,1}^H} \frac{4C}{(1+C)^2 + (\delta/\kappa_{opt}^H)^2}, \quad (F2)$$

where $C = \Omega_c^2/(\kappa_{opt}^L \kappa_{opt}^H)$ is the cooperativity for the coherent coupling between the optical spin-wave mode E_L and the optical spin-wave mode E_H , and $\kappa_{opt}^{L(H)} = \kappa_{opt,0}^{L(H)} + \kappa_{opt,1}^{L(H)}$ is the total emission rate of optical spin-wave mode $E_{L(H)}$. Combined with the previously mentioned conditions, when the atomic number is large enough such that the condition $d_{opt}^{L(H)} \gg 1$ is satisfied, we can approximate that $\kappa_{opt,1}^{L(H)} \gg \kappa_{opt,0}^{L(H)}$, and $\frac{\kappa_{opt,1}^{L(H)}}{\kappa_{opt,0}^{L(H)} + \kappa_{opt,1}^{L(H)}} \simeq 1$. Thus, the total conversion efficiency can reach unity when $C = 1$ and $\delta = 0$. Obviously, this is no fundamental limit of η_{OOC} , which is not the same as MOC.

2. Microwave-to-microwave conversion

When both the incident and converted fields are microwaves, and their wavelengths are larger than the size of the atomic ensemble, the medium for frequency conversion is “superatom-superatom” as shown in Fig. 9, where the wavelength λ_{gr} (λ_{ge}) is larger than the size of atoms, L .

Due to $\lambda_{gr,ge} > L$, we can treat the atomic ensemble as a superatom, as described in Sec. III of the main text. The excitation of state $|r(e)\rangle$ in an N -atom ensemble can be represented by the bosonic operator $S_{L(H)}^\dagger = \frac{1}{\sqrt{N}} \sum_j \sigma_{gr(e),j}^+$ according to the Holstein-Primakoff approximation, where subscript j denotes the j th atom and $\sigma_{gr(e)}^+ = |r(e)\rangle \langle g|$ is the raising operator. Atoms with different excited states ($|r\rangle$ or $|e\rangle$) are regarded as superatoms with different excited states ($|S_L\rangle = S_L^\dagger |G\rangle$ or $|S_H\rangle = S_H^\dagger |G\rangle$), where $|G\rangle$ is the ground state of both superatoms. Since the wavelength of the spontaneously radiated light from the superatom is larger than the atomic-ensemble size, the superatom exhibits an enhanced as

well as uniform radiation rate in all directions, and the total emission rate of the superatom S_L (S_H) is $\gamma_{mw}^L \simeq N\gamma_{gr}$ ($\gamma_{mw}^H \simeq N\gamma_{ge}$). In the single- or multichannel model, the coupling rate between the incident microwave b_{in} (a_{in}) and the superatom S_L (S_H) is $\gamma_{mw,1}^L = \frac{3\lambda_L^2}{4\pi^2 w_{0,L}^2} \gamma_{mw}^L$ ($\gamma_{mw,1}^H = \frac{3\lambda_H^2}{4\pi^2 w_{0,H}^2} \gamma_{mw}^H$), where $w_{0,L(H)}$ is the beam waist of the incident microwave b_{in} (a_{in}). Then the rate of superatomic S_L (S_H) radiation of microwaves into uncollected space is $\gamma_{mw,0}^L = \gamma_{mw}^L - \gamma_{mw,1}^L$ ($\gamma_{mw,0}^H = \gamma_{mw}^H - \gamma_{mw,1}^H$).

Similar to the conversion schematic of MOC introduced in Sec. V of the main text, we can realize MMC by introducing a coherent coupling between the two microwaves, and the corresponding schematic is shown in Fig. 9(b). By a control light field with Rabi frequency Ω_c , the MMC system can be described by the Hamiltonian

$$H = \delta S_H^\dagger S_H + \Omega_c (S_L^\dagger S_H + S_L S_H^\dagger) + [\sqrt{2\gamma_{mw,1}^L} S_L^\dagger b_{in} + \sqrt{2\gamma_{mw,1}^H} S_H^\dagger a_{in} + \text{H.c.}], \quad (F3)$$

where $\delta = \omega_e - \omega_a$ is the detuning between the atomic transition ω_e and the microwave frequency ω_a , where another microwave ω_b is on resonance with the corresponding transition ω_r and the external drive matches the frequency difference between the two microwaves.

The MMC model enables bidirectional frequency conversion between two microwave frequencies and the quantum conversion efficiency is

$$\eta_{MMC} = \frac{\gamma_{mw,1}^L}{\gamma_{mw,0}^L + \gamma_{mw,1}^L} \frac{\gamma_{mw,1}^H}{\gamma_{mw,0}^H + \gamma_{mw,1}^H} \frac{4C}{(1+C)^2 + (\delta/\gamma_{mw}^H)^2}, \quad (F4)$$

where $C = \Omega_c^2/(\gamma_{mw}^L \gamma_{mw}^H)$ is the cooperativity for the coherent coupling between the superatom S_L and the superatom S_H . Similar to the conversion efficiency of MOC, the third term is the internal conversion efficiency, reaching unity when $C = 1$ and $\delta = 0$. The maximum achievable η_{MMC} is intrinsically limited by the extraction factors $\frac{\gamma_{mw,1}^L}{\gamma_{mw,0}^L + \gamma_{mw,1}^L} \frac{\gamma_{mw,1}^H}{\gamma_{mw,0}^H + \gamma_{mw,1}^H}$. Thus, we can obtain

$$\eta_{MMC} \leq \frac{\gamma_{mw,1}^L}{\gamma_{mw}^L} \frac{\gamma_{mw,1}^H}{\gamma_{mw}^H} = \frac{3\lambda_L^2}{4\pi^2 w_{0,L}^2} \frac{3\lambda_H^2}{4\pi^2 w_{0,H}^2}. \quad (F5)$$

According to the diffraction limit of a free-space Gaussian beam ($w_{0,L(H)} > 2\lambda_{L(H)}/\pi$), the upper bound of η_{MMC} is $(3/16)^2$, which is smaller than the η of MOC.

APPENDIX G: THE DISCUSSION FOR NEGLECTING RYDBERG BLOCKADE

Figure 4 presents the theoretical upper limit of the conversion efficiency, obtained under ideal conditions without considering the effect of Rydberg blocking. We need to verify our parameters satisfy both requirements: (1) sufficient atom number for large OD, and (2) negligible Rydberg blockade. Here, we justify that neglecting Rydberg blockade is feasible for two reasons.

(1) Low excitation. For practical quantum applications of microwave-optical conversion, the input microwave signals are typically single photons or weak coherent states.

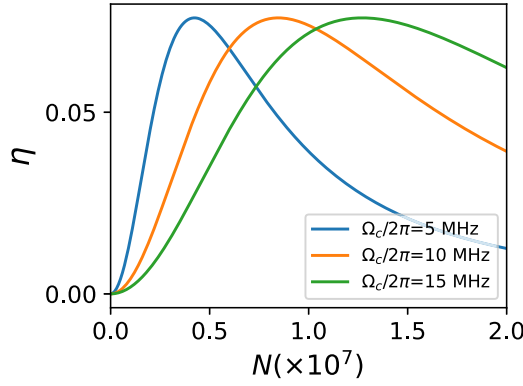


FIG. 10. Conversion efficiency (η) as a function of atom number N for various Rabi frequencies Ω_c ; the optimal number is influenced by Ω_c .

This ensures a mean excitation number in the atomic ensemble at the level of a few or dozens, making blockade effects negligible even for an atomic cloud of the size of millimeters.

For potential quantum application of the microwave-to-optical conversion, the input microwave signal is actually very weak, which leads to very low excitation number in the atomic ensemble; thus, the equivalent excitation number in the ensemble is limited for practical applications.

(2) Low atomic density. According to Ref. [27], a cigar-shaped atomic cloud of size $\sigma_x = \sigma_y = 66 \mu\text{m}$, $\sigma_z = 2.1 \text{ cm}$, has a volume $V = \sqrt{2\pi}^3 \sigma_x \sigma_y \sigma_z$ that can contain the atom number $N > 4.3 \times 10^7$; then mean interatomic spacing is around $2 \mu\text{m}$. Thus, the blockade effect cannot occur when atoms have a Rydberg blocking radius $R < 2 \mu\text{m}$. For Rydberg atoms possessing a larger blocking radius, i.e., the higher the principal quantum number, a smaller Rabi frequency Ω_c of the control light can be used to decrease the optimal atom number N . For example, as shown in Fig. 10, when $\Omega_c/2\pi = 5 \text{ MHz}$, the optimal number is around 4×10^6 . With the same size of cigar-shaped cluster, the optimal number can be realized when $R < 4.4 \mu\text{m}$, which corresponds to a higher principal quantum number. Moreover, a larger volume of clusters also can be used to realize a larger atom number without the influence of the Rydberg blockade.

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