Geometric control of collective spontaneous emission

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We demonstrate quantum control of collective spontaneous emission by fast state-dependent geometric phase patterning. In particular, by driving transition cycles in $^{87}$Rb D1 line with counter-propagating, shaped sub-nanosecond pulse pairs, we temporally control a few-photon D2-excited $^{87}$Rb gas in its directional superradiant states, which allows one to redirect the superradiance with high efficiency on timescales much faster than the emission time itself, and even turn off the collective emission for its recall later. Accompanying the phase writing is efficient optical acceleration of the laser-cooled gas, which is measured to corroborate with our estimation of $\sim 70\%$ control efficiency limited by hyperfine interaction and spontaneous decay. Substantial improvement of the control efficiency is expected with better atomic state preparation and with shorter and more advanced pulse control techniques. Our work represents a first step toward accurate, active control of collective optical dipoles for studying many-body dissipative dynamics of excited gases, as well as for numerous quantum optical applications.

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I. INTRODUCTION

Spontaneous emission is typically a decoherence effect to avoid when levels in small quantum systems are chosen to encode information for e.g., quantum computation, simulation, or sensing [1–4]. On the other hand, spontaneous emission is the process for light-matter quantum information transfer. As “spontaneous” as it is, the information flow during the process can be controlled between long-lived matter degrees of freedom and a pre-aligned single-mode electro-magnetic continuum [5, 6]. In particular, since the seminal work byDicke in 1954 on super- and subradiant effects of light emission by ensembles of excited atoms [7], it is now well-known that the spatio-temporal properties of spontaneous emission are in principle dictated by collective properties of the atoms themselves. For spatially extended atomic ensembles, the timed phase correlations of the collective “spin” excitations (the spin wavevectors) can direct superradiant emission into narrow solid angles [8, 11], suggesting unique opportunities of controlling spontaneous emission for realizing coherent photon-atom interfaces [12, 13]. More recently, it has been realized that these spin wavevectors in atomic arrays open up completely new opportunities within quantum optics, such as to realize waveguiding of light by the array [14–16], atomic mirrors [17, 18], highly subradiant states [19, 20] including emergent Weyl excitation [21] and topological guided edge modes [22, 23]. One major bottleneck to exploring and controlling all of these phenomena is the absence of techniques to control the spatio-temporal properties (e.g., modify the spin wavevector) of the optical excitation, on rapid time scales faster than the typical emission time of atoms themselves.

In this work, we demonstrate quantum control of collective spontaneous emission. Using a dilute laser-cooled atomic sample and a high-speed optical pulse shaping technique, we demonstrate a general method to rapidly and precisely shift the wavevectors of electric dipole collective spin excitations in the time domain, resulting in states with vastly different collective emission characteristics. The rapid $k$–space shift is achieved by geometric phase patterning of the collective spin excitation, through cyclic driving of an auxiliary transition [12] with counter-propagating shaped laser pulses. We use this quantum control to re-direct the forward superradiant emission of an initially prepared spin excitation into a different phase-matched direction with high efficiency ($\sim 70\%$), and furthermore demonstrate the reversible shut-off of the emission for its recall later. The method is readily applicable as well to gases with higher density, or to ordered arrays of atoms with suppressed random scatterings. We thus expect that this state-dependent phase patterning technique contribute to the development of a new class of quantum optical devices [12, 13, 21], and in addition to unlock novel research opportunities for strongly interacting dipolar excited gases [21, 22, 25, 28], by allowing access to low-dissipation subradiant manifolds through active optical control from the far field.

To illustrate the essential physics of the control technique, we start by discussing the collective excitation of $N$ 2-level atoms subjecting to a weak and short plane wave optical pulse referred to as a probe excitation. The resulting quantum state of the excitation can be written in the timed-Dicke state basis [3, 29]: $|\psi_{TD}(k_p)\rangle = S^+ (k_p) |g_1, g_2, \ldots g_N\rangle$ with collective excitation operator

$S^+$.
The spatial and temporal properties of spontaneous emission can be calculated from the electric field operator $\hat{E}(\mathbf{r})$:

$$S^+(\mathbf{k}_p) = \frac{1}{\sqrt{N}} \sum_i e^{i(\mathbf{k}_p \cdot \mathbf{r}_i)} |e_i\rangle \langle g_i|,$$

where $\mathbf{r}_i$ is the location of the $i$th atom in the sample, and $\mathbf{k}_p$ the wavevector of the spin excitation transferred from the probe light. The spatial and temporal properties of spontaneous emission are illustrated with fringes in the atomic sample. The optical control leads to momentum transfer with the probe frequency $\omega$ and the spatial and temporal properties of spontaneous emission.

The enhanced forward detection and the mode profile of the collective emission. For $N \gg 1$ the initial rate of photon emission into the mode is given by

$$\bar{g}(\mathbf{r}) = \sqrt{\frac{\hbar k_p}{8\pi \epsilon_0}} \sqrt{N \sigma_p \Gamma_\theta} \epsilon(\mathbf{r} \cdot \mathbf{k}_p) e^{i(\mathbf{k}_p \cdot \mathbf{r})}.$$

Here $\epsilon(\mathbf{r}) = \frac{1}{N} \int \phi(\mathbf{r}) d\mathbf{r}_p$ is a normalized column density, $\sigma_p = k_p \alpha_i$ is the resonant absorption cross-section, and the imaginary part of the resonant polarizability $\alpha_i$ is related to the dipole element $d_{eg}$ and the single atom spontaneous emission rate $\Gamma$ through $\hbar \Gamma \alpha_i = 2\langle \mathbf{d}_{eg} \rangle^2$. (While $\mathbf{d}_{eg}$ and $\Gamma$ are directly related for two-level atoms, this formula also generalizes to atoms with level-degeneracy.)

Equation (1) provides both the amplitude and the mode profile of the collective emission. For $N \gg 1$ the initial rate of photon emission into the mode is given by $i^{(1)}_p = \frac{2\alpha_p}{\hbar \omega_p} \int |\bar{g}(\mathbf{r})|^2 d^2r_\perp = C_{N,p} \Gamma$, with $C_{N,p} = \frac{N \sigma_p}{8\pi^2} \int g^2(\mathbf{r}_\perp) d^2r_\perp$ related to average optical depth along $\mathbf{k}_p$ as $C_{N,p} \approx \frac{OD_p(r_\perp)}{4}$, $OD_p(r_\perp) = N \theta_c(r_\perp) \sigma_p$ and $OD_p \equiv \int OD^2_p(r_\perp) \int OD_p(r_\perp)$. The enhanced forward emission is associated with the constructive interference.
of light emission from all the atoms in the $k_p$ direction. We further approximate the time-dependence of $\mathbf{r}(r, t)$ (which has a spectrum of $k$-components) with that for the light emission in the $k_p$ direction \[30\] \[33\], and arrive at the time-dependent collective spontaneous emission rate

$$i_p^{(1)}(t) \approx \langle \mathbf{D}^p/4 \rangle \Gamma e^{-(1+i\mathbf{D}^p/4\Gamma)t}.$$  

(2)

Here, the exponential factors of $e^{-\Gamma t}$ and $e^{-i\mathbf{D}^p t/4}$ account for the (non-collective) decay into $4\pi$ and enhanced forward emission along $k_p$, respectively.

We now discuss control of the collective spontaneous emission as schematically shown in Fig. 1. With the $S^+(k_p)$ excitation and after a $\Delta t_1$ delay, an optimally chirped control pulse with wavevector $k_c$, duration $\tau_c$, and its retro-reflection with $-k_c$ and optical delay $\tau_d$ are successively applied to rapidly drive a cyclic transition between $|g\rangle$ and an auxiliary state $|a\rangle$ (returning back to $|g\rangle$) in a quasi-adiabatic and intensity-insensitive manner \[34\]. The geometric phase $\varphi_G(t) = \pi + 2k_e \cdot r$ determined by the optical phases of the pulses is imprinted to $|g\rangle$ for an atom at location $r$, leaving state $|e\rangle$ unaffected either due to selection rules, or by a large $|e\rangle - |a\rangle$ energy difference in which case a large $\omega_p, \omega_c$ difference would enable background-free detection of $\omega_p$ photons.

The ideal state-dependent phase patterning, achievable with strong control at the limit of short $\tau_{c,d}$, can be formally expressed as

$$U_c(\varphi_G) = \prod_{i=1}^{N} \left[ 1 + e^{i\varphi_G(r_i)} - 1 \right] |g_i\rangle \langle g_i|.$$  

(3)

The collective spin excitation is thus shifted to $|\psi_{TD}(k_p)\rangle = U_c |\psi_{TD}(k_p)\rangle = S^+(k_p = k_p - 2k_c)U_c |g_1, g_2, ..., g_N\rangle$ for $\Delta t_1 = 0$. Similar to the forward superradiance, the $S^+(k_s)$ excitation to the phase-patterned ground-state atoms radiates efficiently and collectively if $|k_s\rangle$ matches $|k_p\rangle$, which is achieved experimentally by adjusting $k_c, k_e$ toward an intersection angle $\theta = \cos^{-1}(|k_s|/|k_p|)$. With the perfect phase matched condition, the decays of redirected collective excitation along $k_p$ should follow Eq. (2) in an analogous manner to the forward superradiance.

The state-dependent phase-patterning can be applied multiple times. By applying $U_c$ again after a $\Delta t_2$ delay, we expect collective spin excitation $|\psi_{TD}^\prime\rangle = U_c^\prime |\psi_{TD}^\prime\rangle = S^+(k_s = k_p - 4k_c)U_c^\prime |g_1, g_2, ..., g_N\rangle$, with completely suppressed collective emission since $|k_s\rangle$ is significantly larger than $|k_p\rangle$. Here $|\psi_{TD}^\prime\rangle$ would be a subradiant state with vanishing decay rate if the atomic positions $r_i$ are properly prepared in sub-wavelength lattices \[20\]. In our case, the random positioning results in emission with incoherent phase $\langle \mathbf{r}(r) = 0 \rangle$ at the single-atom decay rate $\Gamma$.

We now discuss implementation of the state-dependent phase-patterning scheme in this work. The probe optical transition is implemented on the $^87\text{Rb}$ $5S_{1/2} - 5P_{3/2}$ D2 line with transition wavelength $\lambda_{D2} = 780$ nm, $k_p = 2\pi/\lambda_{D2}$ and natural linewidth of $\Gamma_{D2} = 2\pi \times 6.07$ MHz. For isolated single atoms, the electric dipole energy decays with time constant $\tau_{D2} = 1/\Gamma_{D2} = 26.2$ ns. The few-photon level collective dipole excitation is induced by a resonant nanosecond laser pulse that couples the hyperfine ground state $5S_{1/2}$ with $F = 2$ and excited state $5P_{3/2}$, $F’ = 3$, represented by $|g\rangle$ and $|e\rangle$ in Fig. 1, respectively. The auxiliary transition is implemented on the D1 line with $|a\rangle$ representing the $5P_{1/2}$ with $F’ = 1, 2$ levels, with $\lambda_{D1} = 795$ nm, $k_c = 2\pi/\lambda_{D1}$ and $\Gamma_{D1} = 2\pi \times 5.75$ MHz.

We choose the polarization for the probe and control lasers to be along $\mathbf{e}_y$ and $\mathbf{e}_x$ respectively. The $U_c$ operation is implemented by cyclically driving the D1 transition with the counter-propagating sub-nanosecond chirped pulses, so as to phase-pattern the $5S_{1/2}$ with the D1 D2 transition frequency difference. Taking the $\mathbf{e}_y$ direction as the quantization axis, the $\pi$–couplings to $5P_{1/2}$ would be with equal strengths and detunings for all the five $5S_{1/2} = 2, m_F$ Zeeman sub-levels, and with vanishing hyperfine Raman coupling, if the $5P_{1/2}$ hyperfine splitting $\Delta m_{1,2} = 2\pi \times 814.5$ MHz can be ignored. The approximation helps us to establish the simple 2-level control picture in Fig. 1 even for the real atom. Practically, the hyperfine dephasing effects can be suppressed for small $\tau_c$, if the optical delay $\tau_d$ can be adjusted to match $2\pi/\Delta m_{1,2} = \approx 1.23$ ns. In this work the delay $\tau_d = 1.36$ ns is fixed by the optical path of a $R = 200$ mm concave retro-reflection mirror outside the vacuum (Fig. 1k), and the dipole control efficiency is correspondingly limited. This limitation can be overcome by preparing atoms in either $|m_F = 2\rangle$ or $|m_F = 0\rangle$ states, or, alternatively, by controlling the multi-level dynamics with advanced multi-pulse control techniques \[35\] \[36\].

With the weak coherent probe, we expect phased electric dipole oscillation by all the atoms \[32\] \[37\] \[38\]. The average collective excitation number is $N\theta_p^2$, with $\theta_p = \frac{1}{2} \pi \Omega_p dt$, with $\Omega_p$ the Rabi frequency of the probe pulse. Equation (2) for the single $S^+(k_p)$ excitation is accordingly modified to describe the $E_c(k_p)$ mode emission in the linear optics regime \[10\] \[11\] \[39\], with photon emission rate $i_p(t) \propto N^2$ as

$$i_p(t) \approx N\theta_p^2(\mathbf{D}^p/4)\Gamma_{D2} e^{-(1+i\mathbf{D}^p/4\Gamma_{D2})t}.$$  

(4)

The non-ideal $U_c$ control in presence of e.g. laser intensity variation or imperfect pulse shaping, shall only partly convert the $S^+(k_p)$ into $S^+(k_s)$ excitation and further induce sub-wavelength density modulation in $\varphi(r)$, thus we expect simultaneous and Bragg-scattering coupled superradiant emission into both the $E_c(k_p)$ and $E_c(k_s)$ modes. Practically for optical control with the focused laser beam as in this work, we numerically find the ground state atoms not shifted in momentum space are often associated with dynamical phase broadening, leading to suppressed $S^+(k_p)$ excitation and distorted sub-wavelength density fringes. Taking into account dipole control indelibility and atom loss by the non-ideal $U_c$ control, the redirected superradiance into the $E_c(k_s)$ mode in absence of
efficient coupling to other decay channels is with a photon emission rate

\[ i_s(t) \approx f_d \frac{OD_s}{OD_p} i_p(\Delta t_1) e^{-\left(1+(1-t)\frac{OD_s}{4}\right)\Gamma_{D2}(t-\Delta t_1)}. \tag{5} \]

Similar to \( \frac{OD_s}{OD_p} \), here the average optical depth \( OD_s \) along \( k_p \) decides the superradiant cooperativity of the unperturbed atomic sample for the \( E_s(k_p) \) mode emission. The dipole control efficiency \( f_d \in [0,1] \) and atomic loss factor \((1-l) \in [0,1]\) can be evaluated in the single atom picture for dilute atomic samples, as in Appendix B. Similar to Eq. \( (2) \), for Eqs. \( (1) \) \( (5) \) the wavefronts for \( E_s(k_p) \), \( E_s(k_v) \) mode emission follows the column density of atomic sample \( \rho_p(r_+), \rho_s(r_-) \) respectively, with \( E_s(k_s) \) expected to subject to residual dynamic phase modulation due to the imbalanced intensity by the retro-reflecting laser beams. Equation \( (5) \) thus serves to characterize the quality of implementing \( U_c \) to control the collective spontaneous emission in this work.

The \( S^+(k'_s) \) excitation in the random atomic gas is expected to decay with a rate close to \( \Gamma_{D2} \). Before complete decay, revival of superradiant emission is expected if a \( U_c^{-1} \) operation can be applied to the \( S^+(k'_s) \) excitation. The \( U_c^{-1} \) operation is ideally achieved by reversing the time-order of the counter-propagating control pulses for studying the \( S^+(k'_s) \) decay dynamics \( \tag{10} \). In this work, \( U_c^{-1} \) is effectively implemented with a state-dependent standing wave diffraction of atoms \( \tag{11} \), so as to verify the reversibility of the collective emission control.

II. EXPERIMENTAL OBSERVATIONS AND ANALYSIS

We prepare \( N \approx 10^4 \) \(^{87}\)Rb atoms in the 5S\(_{1/2}, F=2\) hyperfine level in an optical dipole trap with up to \( \sim 4 \times 10^{12} \text{cm}^{-3} \) peak density and \( T \sim 20 \mu\text{K} \) temperature. After its release from the trap, the dipole excitation is induced by a \( \tau_p = 3 - 6 \) ns, \( I_p \approx 10 \text{mW/cm}^2 \) \( e_p \)-polarized resonant D2 probe pulse. The Gaussian probe beam has a \( w_p \approx 50 \mu\text{m} \) waist, which is much larger than the \( 1/e \) radius of atomic density profile \( \sigma \approx 7 \mu\text{m} \), validating the plane-wave excitation picture. Pairs of \( \tau_c = 0.9 \) ns, \( e_c \)-polarized control pulses, shaped by Rabi frequency \( \Omega_x(t) = \Omega_0 \sin(\pi t/\tau_c) \) and instantaneous detuning \( \delta_c(t) = -\delta_0 \cos(\pi t/\tau_c) \) (from the middle point of the \( F=2 \rightarrow F'=1,2 \) hyperfine lines), are then applied. With \( \approx 20 \text{mW} \) peak power, peak intensity parameter \( s \sim 10^6 \) \( (s \equiv I/I_{s1} \text{ and } I_{s1} = 4.49 \text{mW/cm}^2 \text{ is the D1 transition saturation intensity}) and peak Rabi frequency \( \Omega_0 = \sqrt{s/2\Gamma_{D1}} \) at GHz-level are reached by focusing the \( k_c \)-control beam into a waist of \( w \approx 13 \mu\text{m} \) at the atomic sample. The retro-reflected \(-k_c \) pulse is optically delayed by \( \tau_r = 1.36 \) ns, with slightly increased beam waist \( w_r \) and reduced Rabi frequency, possibly due to wavefront distortion by the vacuum viewport.

Instead of monitoring the forward \( E_s(k_p) \) mode emission \( \tag{10} \) \( \tag{11} \), in this work the redirected \( E_s(k_s) \) mode su-

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2.png}
\caption{Collectively enhanced spontaneous emission under coherent control and monitored by the redirected emission signals. All the signals are histogramed from gated photon counting measurements (time bin \( \delta t=256 \) ps), with \( \tau_p = 3 \) ns and \( \tau_p = 6 \) ns for (i) - (xi) and (xii) - (xiv) respectively. The probe is between \( -\tau_p < t < 0 \). The control pulse is with \( \tau_c = 0.9 \) ns, \( \Omega_0 \approx 2\pi \times 2.7 \) GHz, \( \delta_0 = 2\pi \times 3.4 \) GHz. Curve (i) - (v) gives redirected superradiance with \( \Delta t_1=0.6, 4.6, 8.6, 12.6, 16.6 \) ns. In curve (vii) - (xiv) the superradiance redirected at \( \Delta t_1=0.6 \) ns is turned off at \( \Delta t_2=16.6, 12.6, 8.6, 4.6, 0.6 \) ns. Curve (xvii) - (xix) demonstrate partially recalled superradiance at \( \Delta t_3=0.6, 16.6, 14.6 \) ns with a standing-wave diffraction, after the redirection at \( \Delta t_1 = 0.0 \) ns and turn-off at \( \Delta t_2 = 4.6 \) ns. The top left inset gives the Fourier transform of curve (i). All curves are integrated with \( N_{\text{exp}} = 70000 \) measurements, except curve (vi) which was taken in absence of control pulses with \( N_{\text{exp}} = 30000 \) and increased probe strength. The weak scattering of probe from the nearby vacuum window is also detected during \( -\tau_p < t < 0 \). By avoiding nearby optics and through optical filtering, the control light background is completely suppressed from the signals.
\end{figure}

perradiance is collected by a carefully aligned NA=0.04 objective and detected by a multi-mode fiber coupled single photon counter. To enhance the measurement accuracy for small signals, an optical filter at 780 nm is inserted to block possible fluorescence photons at \( \lambda_c = 795 \) nm. To improve the signal collection rate with our laser-cooling experiments that repeat at sub-Hertz level, we operate multiple excitation-control-measurement cycles with a single atomic sample before next loading. The interval \( T_{\text{rep}} \gg 1/\Gamma_{D2} \) is chosen to ensure independence.
of each measurements. The gradual loss of interacting atoms during the repetitions is carefully characterized for studying number-dependent dynamics.

A. Controlled collective spontaneous emission

Our major experimental achievements in this work can be summarized in Fig. 2 which displays time-dependent emission of redirected superradiance into the phase-matched \( E_s(k_s) \) mode, with optimized D1 controls. Without the control, no discernible background fluorescence signal can be seen, as in curve \((v)\) after the \( N_{\text{exp}} \) measurements that integrate to obtain the histogrammed curves. In contrast, the redirection at various \( \Delta t_1 \) efficiently turns on the superradiant emission as in curve \((i)\) \(-\langle v \rangle\), qualitatively reproducing the prediction of Eq. \((5)\). The turn-on time is approximately 1 ns, which is consistent with the \( \tau_p \) duration of the retro-reflected control pulse during which the \( \varphi_G \)-written \( |g\rangle \) amplitude is restored from \( |a\rangle \). The \( \tau \approx 18 \) ns decay constant associated with Eq. \((6)\) is studied in Sec. II B in detail. The \( f \approx 267 \) MHz oscillations are due to a quantum beat between the D2 collective spontaneous emission from \( F' = 3 \) and those from the off-resonantly excited \( F' = 2 \) hyperfine levels \[12\]. In separate experiments we confirm the suppressed amplitude of quantum beats with increased \( \tau_p \) and reduced probe pulse bandwidth.

The diffraction-limited spatial mode of the redirected superradiance \( E_s(k_s) \) is verified by estimating its divergent angle in the far field at 20 mrad level, which is consistent with the \( \sigma \approx 7 \) \( \mu \)m size of the coherently excited atomic sample at 780 nm, according to Eq. \((1)\). We expect slight distortion of the superradiant wavefront, due to the residual dynamics phase imprinted by the intensity-imbalanced control pulse pair, as discussed in Appendix B. The dynamic phase is estimated at 1 rad level across the atomic sample. To collect the redirected emission with a single mode fiber, such wavefront distortion needs to be characterized and compensated for.

We further proceed with the additional \( U_c \) control to turn off the \( E_s(k_s) \) emission, by applying an additional pair of control pulses at various \( \Delta t_2 \) interval to break the \( |k_s| = |k_p| \) phase-matching condition. The resulting pulsed superradiant signals (with \( \Delta t_1 = 0.6 \) ns) are given in curve \((vi)\) \(-\langle xiv \rangle\) of Fig. 2. Similar to previous curves, here the \( \sim 1 \) ns turn-off time reflects the gradual attenuation of the \( |g\rangle \) amplitude by the first part of this control pulse pair.

With the additional \( \varphi_G \)-written \( |g\rangle \) amplitude restored again from \( |a\rangle \) by the second pulse of the 2nd control, we expect that a superradiance-free \( S^+(k_s') \) excitation is stored into our atomic sample. A full investigation of such collective excitation by implementing a \( U_{-1} \) control is an on-going effort with a modified setup, and will be for a future publication \[10\]. Here we confirm the coherent nature of the turn-off operation after \( \Delta t_3 \) delay, by applying a \( \tau_{\text{KD}} = 10 \) ns, detuning \( \Delta_{\text{KD}} = -2\pi \times 6 \) GHz, Rabi frequency \( \Omega_{\text{KD}}(t) = \Omega_{\text{KD,M}} \sin(\pi t/\tau_{\text{KD}}) \) shaped control pulse to form a standing-wave lattice along \( e_z \) for the \( F = 2 \) atoms in \( |g\rangle \). The peak Rabi frequency is \( \Omega_{\text{KD,M}} \approx 2\pi \times 2 \) GHz. A Kapitza-Dirac diffraction \[11\] leads to state-dependent sinusoidal dynamic phase modulation \( \varphi_{\text{KD}}(t) = \int t \delta_{\text{KD}}(\tau) \sin^2(k_s \cdot r) d\tau \) to \( |g\rangle \), with light shift \( \delta_{\text{KD}}(t) \approx \Omega_{\text{KD}}(t)^2/\Delta_{\text{KD}} \), that partially converts the phased excitation associated with \( S^+(k_s') \) back to those associated with \( S^+(k_s) \). The conversion efficiency is peaked at local-intensity-dependent time \( t \) and we expect inhomogeneously broadened revival dynamics. As in curve \((xii)\) \(-\langle xiv \rangle\), we observe revived superradiant emission after various \( \Delta t_3 \) in absence of control pulse background, with the turn-on time, amplitude, and duration of the rapidly dephasing revivals agreeing quantitatively with accurate numerical modeling detailed in Appendix B. The observation confirms the reversibility of the \( U_c(\varphi_G) \) operation for super- and sub-radiant state conversions.

B. Decay dynamics

The detection of redirected superradiant emission, as in Fig. 2 is essentially background-free and enables us to precisely study the decay dynamics of the collective excitation under various conditions. As an example, here we study the simplest case of collective decay in a dilute atomic gas, which is fairly accurately modeled with Eqs. \((4)(5)\). Experimentally, we vary the atom number \( N \) for samples loaded into the same dipole trap with nearly identical spatial distribution. The \( E_s(k_s') \) emission at the \( \Delta t_1 = 0.2 \) ns delay is recorded as in Fig. 3. The time-dependent photon emission rate \( i_s(t) \), obtained by normalizing the fluorescence counts with \( N_{\text{exp}} \), counter-time-bin \( \delta t \), and an overall detection quantum efficiency \( Q \approx 0.15 \), nicely follows exponential decay curves for the accessed \( N \) between \( 2 \times 10^3 \) and \( 9 \times 10^3 \) in this work. We extract both the peak emission rate \( i_{\text{max}}N \) and collective decay rate \( \Gamma_N \) with exponential fits, and to study both quantities as a function of atom number \( N \).

The cooperative nature of the collective emission is clearly demonstrated with the \( i_{\text{max}}N \propto N^2 \) relation as in Fig. 3. According to Eqs. \((1)(3)\), the peak collective photon emission rate is given by \( i_{\text{max}}N \approx f_d N C_{N,S} \theta_{D_2}^2 \Gamma_N \) with \( C_{N,S} = OD_s/4 \). We experimentally estimate the average optical depth as \( OD_s = (OD_s^2)^{\beta/2} \), \( \langle \langle \beta \rangle \rangle \) as spatial average, and \( OD_s(\xi) \) the 2D resonant optical depth profile of the atomic sample to be measured along the \( k_s \) direction. In this work, since the \( k_s \) direction is not accessible by our imaging optics, we estimate \( OD_s^2 \) with \( OD_s(\xi) \) measurements (Fig. 5 insets, see Appendix A4 for imaging details.). We have \( OD_s = \xi \times OD_{z_s} \) with \( \xi \approx 0.8 \) to account for the ratio of optical depth integrated along the \( k_s \) and \( e_z \) directions respectively. By comparing the quadratic fit that gives \( i_{\text{max}}N \approx 6 \times 10^{-3} N^{-1} T_{D_2} OD_s/4 \) in Fig. 3b with Eqs. \((1)(3)\), we find \( \theta_p \approx 2 \times 10^{-2} \) for \( f_d = 0.7 \).
We now discuss the decay rate $\Gamma_N$ of the collective emission, which is approximated in Eqs. (2) with the decay of the corresponding timed Dicke state. To make a precise comparison with the theoretical picture, the same data in Fig. 3b is plotted along different atom numbers, with resonant optical depth images along $\psi$, (insets) inferred from absorption images taken at the corresponding experimental conditions. The exponential fit gives the peak count rate $\Gamma_{\text{max,N}}$ and the collective decay rate $\Gamma_N$. (b) $\Gamma_{\text{max,N}}$ and $\Gamma_N$ are plotted vs estimated atom number $N$ and average optical depth $\bar{OD_x}$. The error bars represent the statistical and fit uncertainties of the data.

(See Sec. [1D]). This $\theta_p$ is consistent with the expected excitation by the peak $s \approx 1$, $\tau_p = 5$ ns probe in these measurements [43], considering the large uncertainty in the absolute intensity parameter estimations.

We now discuss the decay rate $\Gamma_N$ of the collective emission, which is approximated in Eqs. (2) with the decay of the corresponding timed Dicke state. Similar to previous studies of forward superradiance [10, 11], here $\Gamma_N$ for the redirected superradiance is also found to depend linearly with $N$, as in Fig. 3b, which is expected since the cooperative enhancement of directional emission shares the same underlying coupled dipole dynamics. To make a precise comparison with the theoretical picture, the same data in Fig. 3b is plotted vs in situ measured average optical depth $\bar{OD_x}$. From Fig. 3b we have $\Gamma_N/\Gamma_{D2} \approx 1.1 + 0.26 \bar{OD}_x$. Using Eq. (5), again with $\xi = \bar{OD}_x/\bar{OD}_D \approx 0.8$ and by taking into account $(1 - l) \approx 0.9$ in these measurements, as discussed in Sec. [1D], we obtain $\Gamma_N \approx (1 l + \nu (1 l) \bar{OD}_D) \Gamma_{D2}$ with $\nu = 0.35 \pm 0.1$, with no freely adjustable parameter but with an uncertainty limited by the $\bar{OD}_x$ estimation in this work. The likely discrepancy between this result and the $\nu = 0.25$, $\Gamma_N/\Gamma = 1 + \bar{OD}/4$ prediction on the collective decay of the timed Dicke state [30][32][44] can be expected, since the measured collective emission $i_s(t)$ is integrated over the $\sigma$-limited solid angle beyond the “exact” $k_s = k_p - 2k_p$ phase matching condition, while the small angle scattering of $E_s(k_s)$ by the sample itself [33] generally affects the collective emission dynamics. A detailed study on the subtle effect enabled by the background-free technique will be for a future work.

C. Optical acceleration

Accompanying the dipole control is optical acceleration of atomic population in the phase-patterned $|g\rangle$ states by the geometric force [45]. The momentum transfer along the control beam with $k = k_p e_z$ can be evaluated by integrating $\langle \hat{F}_z \rangle$ with the single-atom force operator $\hat{F}_z = -\frac{\hbar}{2} \partial_x \Omega_x |a\rangle \langle g | + h.c.,$ as the projected atomic state evolves on the $\{|g\} - \{|a\} \rangle$ Bloch sphere (Fig. 1b). For ideal population inversions, the integrated Berry curvature [46] gives the exact photon recoil momentum $\Delta P = 2\hbar k$, with $\hbar$ the reduced Planck constant. The same effect can also be understood as arising from the phase writing $U_c(r)$ to delocalized matter-waves of atoms in $|g\rangle$ [47], or, in a full quantum picture, as due to quantized momentum exchange between light and matter. As studied in pioneering work by Metcalf and co-workers [31][48], the cyclic rapid adiabatic passage is a robust way to generate strong optical forces, with important applications in laser cooling and more generally in controlling external motion of atoms and molecules [43][50].

We measure the recoil momentum transfer $\Delta P$ by the same D1 chirped pulse pair for the collective dipole control. The velocity change is obtained by fitting the central position shift in calibrated absorption images of atomic sample, after a free-flight with $T_{\text{rep}} = 400 \mu$s, with and without the control as detailed in Appendix A.3. Keeping in mind the Doppler effects due to the acceleration affect negligibly the nanosecond control dynamics, we repeat the control sequence 5 times to enhance the measurement sensitivity. The period $T_{\text{rep}} = 440 \mu$s $\gg 1/\Gamma_{D1}$ is again set to ensure independent interactions. In Fig. 4b, the retrieved $\Delta P$ per control sequence is plotted vs intensity parameter $\sqrt{s}$, for shaped pulses with different $\delta_0$. For controls with nearly zero chirp ($\delta_0 = \pi \times 0.9, 1.0 \text{ GHz}$), $\Delta P$ displays a damped oscillation, which is due to optical Rabi oscillation with broadened periodicity associated with intensity inhomogeneity of the focused laser. The oscillation is suppressed at large $\delta_0$, with $\Delta P$ reaching $80(4)\%$ of the $\sim 2\hbar k$, limit at large $s$. The features of the measurements at vari-
ous control parameters are well reproduced by numerical simulations, as in Fig. 2, and discussed in Sec. II D.

D. Control efficiency: calibration and optimization

To quantify the non-ideal $U_c$ operation implemented in the experiments, we need to properly model the dissipative dynamics of collective dipoles. For this purpose, we introduce the coherent dipole control efficiency, $f_d = \langle \rho_s \rangle = \rho_0 S^+ (k_j) S^- (k_j) \rho_0 S^+ (k_j) S^- (k_j)$, with $\rho_0$, $\rho_s$, the density matrix that describes the weakly D2 excited atomic sample subjected to the non-ideal $\hat{U}_c$ and the ideal, instantaneous $U_c$ control by Eq. (3) respectively. Here $\hat{U}_c(\Omega_0, \delta_0, \eta)$ due to the nanosecond shaped pulse control is parametrized by $\Omega_0$, $\delta_0$, as well as additional laser intensity profile factors $\eta_{1.2}$ for the forward and retro-reflected pulses. The $\eta_{1.2}$ are determined by the location of the atoms in the Gaussian beam profiles and $f_d$ is averaged over the intensity distribution.

Experimentally, we scan the control pulse shaping parameters $\sqrt{\xi} \propto \Omega_0$ and $\delta_0$ for maximal redirected superradiant emission. The data in Fig. 3 are total counts of the redirected spontaneous emission integrated over the time-dependent signal similar to curve (i) in Fig. 2. With consistent atomic samples the collective decay lifetime as detailed in Sec. II B is verified to be quite insensitive to the control quality, confirming the approximate validity of Eq. (6) that relates $f_d$ with the measured superradiant emission rate $i_s(t)$ for both good and bad controls. The data in Fig. 3 thus quite linearly reflects the collective dipole control efficiency $f_d$ and we are able to locate optimal pulse shaping parameters $\Omega_0 = 2\pi \times 2.7$ GHz and $\delta_0 = 2\pi \times 3.4$ GHz that maximize the superradiance redirection by the $\tau_c = 0.9$ ns chirped-sine pulses in these experiments.

To calibrate $f_d$, however, it is insufficient to use Eq. (5) directly due to large uncertainties in estimating experimental parameters such as $\theta_p$, $\bar{OD}_s$, and quantum efficiency $Q$. Instead, we calibrate the control efficiency by accurate modeling based on single-atom dynamics that numerically reproduces the features of both the acceleration and superradiance measurements. In particular, we adjust parameters in numerical simulations so as to optimally match the simulated average momentum shift $\langle \Delta P \rangle_\eta$ in Fig. 3 with experimental results in Fig. 3. The corresponding $f_d$ under nearly identical experimental conditions are then calculated as in Fig. 4. The fairly nice match between the superradiance measurements in Fig. 4(c) and Fig. 4(d) is achieved by uniformly normalizing the total counts in Fig. 4(c), with no additionally adjusted parameters. The remaining discrepancy could be due to breakdown of the smooth $\rho(r)$ assumption leading to Eq. (5) at low $f_d$, and also deviations of actual pulse shape from the chirped sine form, which introduces distortion to the signals for low-$f_d$ parameter regime unstable against its variations. Near the optimal control regime, the simulation suggests we have reached a collective dipole control efficiency $f_d \approx 72 \pm 4\%$, accompanied with the observed $f_d \equiv \langle \Delta P \rangle / 2\hbar k_c = 89 \pm 4\%$ acceleration efficiency. Constrained by the absolute acceleration measurements, we found this optimal $f_d$ estimation to be quite robust in numerical modeling when small pulse shaping imperfections are introduced.

The optimal $f_d$ is limited by $m_F$-dependent hyperfine phase shifts and D1+D2 spontaneous decays during the $\tau_d + \tau_\gamma = 2.26$ ns control. In particular, a $l \sim 10\%$ atom loss due to D1 spontaneous emission and $5P_{1/2}$ population trapping (particularly for $|m_F| = 1$ states) is expected to reduce the number of atoms participating in the D2 collective emission. With atoms prepared in a single $m_F = 0$ state, spontaneous emission limited dipole control efficiency of $f_d = 87\%$, accompanied with an acceleration efficiency $f_d = 97\%$ should be to reachable [Figs. 2(b)(d)] with the same control pulses. Dipole control fidelity approaching 99% is possible by further reducing the $\tau_c$ control time with more powerful lasers and more advanced pulse shaping techniques, as discussed in Sec. III A.
III. DISCUSSIONS

The fast state-dependent geometric phase patterning demonstrated in this work is a general method to precisely control collective dipole excitation and highly directional superradiant emission in the time domain \([12, 51, 53]\). The control is itself a single-body technique, which can be accurately modeled for dilute atomic gases when the competing resonant dipole couplings can be ignored during the pulse duration. As discussed in Appendix \(B\) we emphasize that with increased \(\Omega\) strength and reduced \(\tau_c\) time, it is generally possible to suppress the effects of atom-atom interaction so as to maintain the control precision enabled by the single-body simplicity. With the geometric phase inherited from the optical phases of the control laser beams, it is straightforward to design \(\varphi_G\) beyond the linear phase used in this work and to manipulate the collective spin excitation in complex ways tailored by the control beam wavefronts.

In the following we discuss prospects and technical requirements for controlling high-density gases beyond this experimental work. We then briefly discuss the role of atomic motion, and finally summarize this work.

A. Toward perfect control at short pulse limit

The state-dependent geometric phase patterning \(U_c(\varphi_G)\) in this work is subjected to various imperfections. At the single-body level, the pulse shaping errors combined with laser intensity variations lead to imperfect population inversions and reduced operation fidelity. The imbalanced beam pair intensities lead to spatially dependent residual dynamical phase writing and distortion of the collective emission mode profiles. The hyper-fine coupling of the electronically excited states may lead to inhomogeneous phase broadening as well as hyper-fine Raman couplings, resulting in coherence and population losses as in this work. Finally, the spontaneous decays on both the D1 control and D2 probe channels limit the efficiency of the finite-duration pulse control. However, the imperfections of the control stemming from the single-atom effects are generally manageable with better quantum control techniques \([30, 51, 53]\) well-developed in other fields, if they can be implemented in the optical domain with reliable pulse shaping system of sufficient precision, bandwidth and output power.

In Appendix \(B\) we take one step further and discuss the scaling of the control error \(\delta \varphi\) due to atom-atom interactions in a dense atomic sample, for \(U_c\) that has been nearly perfected for single-body control. In the worst case scenario and with clever choice of \(\mathbf{k}_c\) direction for non-isotropic samples, we suggest the control error for single photon excitation is bounded by \(\delta \varphi_M \sim \sqrt{\delta_N^2 + \Gamma_N^2} / 4 \tau_c\), with \(\delta_N\), \(\Gamma_N\) the largest collective Lamb shift \([30]\) and collective decay rate of the sample under consideration. Thus it is realistic to consider that with a 10-fold reduction of \(\tau_c + \tau_d\) to 100 ps level, the technique may be applied to scenarios as envisioned in refs. \([17, 18, 57]\) on electric dipole transitions with high precision, with various applications envisaged in the field of quantum optics \([12, 13, 24]\), and to unlock non-trivial many-body physics of dipolar interacting gases \([19, 23, 25, 28]\) inaccessible with far-field linear excitation.

For the error-resilient shaped optical pulse control, the 10-fold reduction of control resonant dipole coupling bandwidth and a 100-fold increase of laser intensity, according to discussions in Appendix \(B\). Starting from the sub-nanosecond pulse shaping technique in this work detailed in Appendix \(A\) the improvement is technically challenging but not formidable. To achieve precise control at even shorter time scales, the control pulses may be generated with mode-locked lasers \([49, 58, 61]\) with orders of magnitude enhanced peak power and pulse bandwidth. This prospect may require further developments of precise ultra-fast pulse shaping technology with mode-locked lasers \([62]\).

B. Atomic motion

This work brings together two seemingly unrelated phenomena: The control of collective dipole radiation, and the acceleration of the free emitters. The physics behind the picture is quantum mechanics, which requires state-dependent acceleration during the sub-wavelength scale electric dipole phase control. To avoid the apparent acceleration of ground state atoms, the phase patterning could be chosen to address instead the much less populated state \(|e\rangle\), with excited state couplings \([63]\). However, in either case, the state-dependent acceleration introduces Doppler phase broadening to the controlled dipoles, that limits the coherence time of the collective excitation to \(\tau_D \sim t_c / v_k\) where \(t_c\) is the thermal deBroglie wavelength of the atomic sample and \(v_k = \hbar k / m\) is the recoil velocity associated with the \(S^+(\mathbf{k})\) collective excitation. In this work, with \(t_c \approx 100\) nm for \(^{87}\text{Rb}\) at 20 \(\mu\text{K}\) and \(v_k \approx 6\) mm/s, the Doppler dephasing time \(\tau_D \sim 15\) \(\mu\text{s}\) does not affect the observed superradiance dynamics at the \(\tau_D / 2\) time scale. To suppress the acceleration so as to maintain the phase coherence of the long-lived subradiant excitation in future work, particularly for lighter atoms with larger \(v_k\), or for narrower line transitions with smaller \(\Gamma\) \([59]\), the atoms should be confined by optical lattices in the recoilless or Lamb-Dicke regime, with the optical lattices at a “magic wavelength” and nullified dipole transition frequency shifts.

C. Summary

In this work we experimentally demonstrate and systematically study a state-dependent geometric phase patterning technique that precisely controls collective spon-
taneous emission on an electric dipole transition. The work also includes a careful investigation of $\Gamma_N/\Gamma = 1 + \frac{OD}{4}$ relation for the collective emission enabled by the control technique, and a combined study of optical acceleration during the dipole control. We have provided a first theoretical analysis of this spontaneous emission control. We expect continuation of this work on both experimental and theoretical sides on precise control of dipolar excited high density gases, which should have applications within quantum optics and many-body physics. On the laser technology side, we hope this work motivates additional developments of continuous and ultrafast pulse shaping methods for optimal quantum control of optical electric dipoles.

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Appendix A: Experimental details

1. High speed pulse shaping system

We develop a high speed pulse shaping system to generate both the sub-nanosecond D1 control pulse and nanosecond D2 probe pulse in this work. The system is based on fiber-based electric-optical modulation (fEOM) of an optically amplified external cavity diode laser (ECDL) output [64], followed by a grating based optical filter. A simplified schematic setup is given in Fig. 5.

For the D1 control pulse generation, the ECDL is offset-locked to the hyperfine crossover between the $F = 2 - F' = 1$ and $F = 2 - F' = 2$ transitions of the $^{87}$Rb D1 line. The 30 mW output of the laser is amplified by a Tapered Amplifier (TA) to about 1.8 W. Pulsed diffraction output from an Acoustic-Optical Modulator (AOM) is coupled into fEOM for wide-band microwave modulation driven by a high-speed arbitrary waveform generator (Keysight M8195A), referred to as microwave AWG in the following. With AOM diffraction kept at a low duty cycle, the average laser power coupled into fEOM is kept below 20 mW to avoid photo-refractive damage [65].

To shape the optical pulses with microwave pulses, we use the side-band modulation technique by encoding the pulse shape information into amplitude $A(t) \in [0, 1]$ and phase $\varphi(t)$ of carrier modulation with $\omega_M$ angular frequency. The input-output relation for the complex electric field of the optical wave can be expressed as:

$$E_{out} = e^{i\theta_0 A(t) \sin(\omega_M t + \varphi(t))} E_{in} = \sum_n i^n J_n(\theta_0 A(t)) e^{i n \omega_M t + \varphi(t)} E_{in}. \quad (A1)$$

The phase modulation depth factor $\theta_0$ is decided by the microwave power and fEOM modulation efficiency.

The 2nd line of Eq. (A1) suggests we can simultaneously shape the amplitude and phase of $n^{th}$-sideband with the single fEOM modulation. We choose $n = 3$, $\omega_M = 2\pi \times 16$ GHz, and adjust the microwave power toward $\theta_0 \approx 4$ so as to maximize the modulation efficiency for the sideband decided by the Bessel function $J_3(\theta_0)$. To suppress the unwanted sidebands, collimated fEOM output is sent through a $\sim 13$ GHz bandwidth optical filter, which is composed of a diffraction grating (2400 lines/mm) and a single mode fiber. With the modulation efficiency limited by $|J_3(\theta_0)|^2 \sim 0.18$ and after all the coupling losses, we achieve 20 mW peak power for the $n = 3$ order sideband with modulation bandwidth limited by the grating filter. Due to the large $\omega_M$, the $n = 2, 4$ sidebands are less than 30% of energy for typical pulse shapes [Figs. 5(a)(c)]. The on-off power ratio is about 400 : 1. The whole system is referred to as our D1 optical AWG.

In this work the D1 optical AWG serves to generate the sub-nanosecond chirped-sine pulses as in Fig. 1 in the main text. Distortion of the actual pulse shape is expected due to the limited optical and microwave bandwidth and nonlinearity of the whole modulation system. Therefore, instead of assuming programmed pulse shapes, we directly measure the optical waveform to confirm its functional form, by beating the pulsed output with a reference CW laser which is $\Delta_0 = 2\pi \times 4.6$ GHz blue-detuned from the $F = 2 - F' = 1, 2$ crossover of the $^{87}$Rb D1 line. Typical intensity and beat note measurements are shown in Fig. 6. The intensity measurements in Figs. 6(a)(c) deviate from the $\sin^2(\pi t/\tau_c)$ model programmed for the $3^{rd}$ sideband, mainly because of the unwanted sidebands (mainly the n=2 and n=4 orders) not fully suppressed by the grating filter. However, we expect the unwanted sidebands affect negligibly the D1 control due to their $\sim 16$ GHz or larger detunings from the atomic resonance. The beat note signals as in Figs. 6(b)(d) are quite well fitted by the interference expected from the chirped-sine form as $\sin(\pi t/\tau_c) \sin(\Delta_0 - \delta_0 \sin(\pi t/\tau_c)/\pi + \varphi_0)$. The fitted $\delta_0$ that enters the discussion in this work in the main text are generally smaller than their programmed values. In addition to the $\delta_0$ calibration, we also calibrate the linearity of $\sqrt{s}$ with respect to the programmed values according to the beat note measurements. An over-
all absolute value correction factor $\kappa$ multiplied to $\sqrt{s}$ is instead estimated by comparing simulation with experimental measurements of optical acceleration, as detailed in Appendix B.3.

Additional pulse distortion could come from possible sub-pulses due to multiple reflections at interfaces for the microwave and optical pulse propagation. To suppress electronically generated sub-pulses, care is taken to choose microwave cables with minimal lengths in this work. To suppress optically generated sub-pulses, the optical elements following the $\text{fEOM}$ output are chosen to minimize unwanted retro-reflections. The absence of unwanted sub-pulses in the $\text{k}_c$ beam is verified for delay beyond 1 ns and for relative intensity beyond -40 dB, with a multi-mode fiber coupled photon counter at nanosecond absolute time resolution. Within one nanosecond delay, any sub-pulse would lead to distortion of the nanosecond pulse shape itself. However, it is difficult to tell whether the small distortion as in Fig. 6 is indeed due to pulse distortion, or due to finite response of our fast photo detector (Thorlabs PDA8GS). By combining all the pulse measurements with an overall setup analysis, we conclude that any sub-pulse co-propagating with the main pulse in our system is below 35 dB in relative intensity.

The nanosecond D2 probe pulse is generated by another ECDL-$\text{fEOM}$ setup that shares the same microwave AWG. The D2 optical AWG system also serves to generate the cooling laser $[66]$. To ensure plenty of output power, the D2 laser is modulated by the $\text{fEOM}$ before being amplified. This reversion of setup order introduces extra nonlinearity by the TA, leading to imperfect sine wave distortion, or due to finite response of our fast photo detector (Thorlabs PDA8GS). By combining all the pulse measurements with an overall setup analysis, we conclude that any sub-pulse co-propagating with the main pulse in our system is below 35 dB in relative intensity.

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2. Experimental Sequence

To produce the atomic sample in this work, up to $\sim 10^7$ atoms are loaded into a magneto-optical trap (MOT) in less than 1 sec. Assisted by polarization gradient cooling, up to $10^5$ atoms are then loaded into a 1064 nm crossed optical dipole trap at $\sim 0.5$ mK trap depth, which are subsequently transferred to a 840 nm dipole trap with up to $2 \times 10^4$ atoms. This system is designed for evaporation of the sample to quantum degeneracy $[67]$. In this work, slight evaporation in the hybrid trap produces the $\sim 20 \mu$K atomic sample with up to $4 \times 10^{12}$ cm$^{-3}$ peak density. By adjusting the power ratio of the dipole and dipole traps, the aspect ratio of the atomic sample can be tuned for different measurements, and are estimated with both imaging along the x direction, and an auxiliary $z$-imaging path with removed retro-reflecting mirror. In particular, the measurement results presented in Fig. 8 are with approximate Gaussian radius of $(1.1, 1.0, 0.9) \times \sigma$ with $\sigma = 8 \mu$m along the $x, y, z$ axis respectively. In other measurements we have slightly elongated samples along $z$ and with $\sigma \sim 7 \mu$m along the minor axes.

Up to $N_{\text{rep}} \approx 100$ cycles of the probe-control sequence are applied to the atomic sample right after its release from the optical trap. Within each cycle, the probe pulse with central frequency resonant to $5S_{1/2}F - 5P_{3/2}F' = 3$, and then the sequence of the D1 control pulses, are applied to the atomic sample. Synchronized with the probe pulse is an electronic trigger that starts a photon-counter to record the redirected fluorescence photons. We adjust the probe excitation strength $\theta_p$ so that typical counting probability per cycle $p \sim 0.1 - 0.3$ is small enough to avoid counter saturation. After each probe-control-measure cycle and a $\sim 100$ ns delay, a 100 ns repumping pulse resonant to $F - F' = 2$ is applied to repump atoms in $F = 1$ to $F = 2$, with efficiency estimated to be better than 85%, before the next cycle.

The repeated superradiance measurements to the same atomic sample are accompanied by heating and loss of atoms that contribute to reduced collective emission signals. By comparing the intensity of superradiance for different repetition $N_{\text{rep}}$, we found the superradiant intensity decreases to $\sim \{30\%, 80\%, 95\%\}$ with $N_{\text{rep}} = \{100, 50, 10\}$ respectively. To find a balance between signal-to-noise and the atom-loss errors, for most of experiments in this work we only extract and average the signals from the $N_{\text{rep}} = 50$ repetitions of measurements, except for the density effect investigation as in Sec. II.B where the average is limited to the first $N_{\text{rep}} = 10$ cycles. By further repeating the $N_{\text{rep}}$-cycles $N_c$ cold atom loading times, the overall measurement repetition is given by $N_{\text{exp}} = N_c N_{\text{rep}}$. 
3. Absorption imaging for optical acceleration

In optical acceleration measurements, we remove the probe and repumping pulses, and repeat the control cycle five times for good acceleration but negligible heating. The central position of the atomic samples after a $\tau_{\text{tot}} = 400$ $\mu$s time of flight (TOF) is estimated with Gaussian fit of calibrated absorption images, as detailed by the caption of Fig. 7.

4. Resonant OD and atom number measurements

The absorption imaging setup as schematically illustrated in Fig. 7 not only helps us to quantify the optical acceleration effect with TOF technique, but also to directly measure the optical depth profile $OD_x(y, z)$ and atom number $N$ as in Sec. II B. To investigate the $\Gamma_3/N = 1 + OD/4$ relation, extra care was taken to extract the $OD_x(y, z)$ images from the resonant absorption images. Here $OD_x(y, z)$ to be measured should be the unpolarized atoms in the weak excitation limit, with in situ $\phi(r)$ distribution close to those in the quantum optics experiments and for both low $OD < 1$ and quite high $OD \sim 3.5$. To ensure consistent $\phi(r)$ distribution to be measured, a short exposure time of 20 $\mu$s is chosen. To collect sufficient counts on the camera, we use imaging beams with quite high intensity in the range of $I_0 = 1 \sim 20$ mW/cm$^2$ and thus with a saturation parameter $s = 0.3 \sim 7$ assuming $I_s = 3.05$ mW/cm$^2$ [68] for $\pi$ transition of $5S_{1/2}F = 2 - 5P_{3/2}F' = 3$. We reduce the measurement uncertainty related to saturation effects following techniques similar to refs. [69, 70]. In addition, to avoid measurement uncertainty related to low local counts for the highly absorbing samples, we calibrate the peak $OD$ of the in situ samples with time-of-flight (TOF) images at reduced $OD$. The processes are detailed as following.

We start by repeated absorption imaging measurements for nearly identical TOF samples with 2D transmission profile $T(I) = I/I_0 > 75\%$, with incoming $I_0(y, z)$ and transmitted $I(y, z)$ intensities recorded on the camera. The optical depth profile in the weak excitation limit can be approximately as $OD_x(y, z) = -\log(T(I) + (I_0 - I)/I_s^{\text{eff}} [69, 70])$. Here $I_s^{\text{eff}}$ is an effective parameter for calibrating our saturation intensity measurements. By globally adjusting $I_s^{\text{eff}}$ and thus the $(I_0 - I)/I_s^{\text{eff}}$ term, we obtain consistent $OD_x(y, z)$ from all the measurements with $I_0 = 1 \sim 20$ mW/cm$^2$ with minimal variations. Notice that the radiation pressure during the imaging process does not significantly vary the power-broadened atomic response.

The optimally adjusted $I_s^{\text{eff}}$ serves to extract the $OD_x(y, z)$ spatial profile for atomic sample immediately after their release from the dipole trap, as in Fig. 3 with approximately identical spatial profiles. In addition, under the consistent atomic sample preparation conditions we also measure the optical depth profile $OD_x^\prime(y, z)$ and total atom number after a $430$ $\mu$s time-of-flight. The time-of-flight greatly reduces the peak linear absorption for the highest $OD$ sample here from the expected $95\% \sim 99\%$ level down to $15\% \sim 25\%$, leading to more accurate estimation of integrated $OD$ that is served to calibrate the in situ $OD_x$ measurements. To account for optical pumping effects that tend to increase the $F = 2 - F' = 3$ light-atom coupling strengths, a factor of 0.85 [68] is multiplied to the extracted $OD_x(y, z)$.

We finally adjust $OD_x$ due to the imaging laser frequency noise in this work by up to $30\%$, according to the measured linewidth broadening of the TOF sample ab-

![Figure 6](image-url)

**FIG. 6.** Intensity and beat note measurements of chirped pulses programmed with $s(t) \sim \sin^2(\pi t/\tau_r)$ and $\delta(t) = -\delta_0 \cos(\pi t/\tau_s)$. (a) Intensity measurement (red points) and $\sin^2$-model (blue line) of resonant pulse with $\tau_s = 0.9$ ns. The model is for the desired $3^{rd}$ sideband of the EOM output. The difference between the measurement and the model is mainly due to the $n = 2, 4$ sidebands detected by the photo-diode. (b) Beat signal (red points) and chirped-sine model fit (blue line) of the same pulse as in (a). The reference CW laser is 4.6 GHz blue detuned. The pulse is programmed with $\delta_0 = 0$ GHz, but the fit suggests $\delta_0 = 2\pi \times 0.1$ GHz, likely developed during the dispersive propagation of the pulse within the optical AWG. Similar data and analysis are presented in (c) and (d) for a chirped pulse with duration $\tau_s = 0.9$ ns, with $\delta_0$ programmed to be $2\pi \times 4$ GHz but $\delta_0 = 2\pi \times 3.4$ GHz according to the fit. The intensity measurements in (a) and (c) are averaged for 4000 times, while the beat note signals in (b) and (d) are single-shot measurements.
A full theoretical description of the light-atom interaction in this work involves dynamics of the fairly densely packed multi-level atoms under the D2 weak excitation and D1 quasi-adiabatic control, with long-range interaction mediated by resonant exchange of photons. The full solution of such a multi-level, many-atom system remains an open theoretical and numerical challenge within quantum optics, which goes beyond the scope of this work. Instead, we focus on the weak D2 excitation and to derive Eqs. (1), (2), (4) in the main text using the ”spin-model” as in ref. [20] for 2-level atoms. We then discuss the fast D1 optical control with a 3-level model by treating atom-atom interactions as perturbations to the single-body dynamics during the control interval, and also to discuss Eq. (5) in the main text. We briefly discuss the quasi-adiabatic control technique leading to Eq. (3) including geometric and dynamic phases, and refer readers to refs. [34, 46, 71, 72] on the control technique and its geometric aspects. Finally we discuss the single-body simulation of the collective dynamics that helps to quantify the experimental observations in this work.

1. Collective spontaneous emission from a dilute gas of 2-level atoms

We consider the interaction between N 2-level atoms with resonant electro-magnetic field at wavelength $\lambda_p$ and frequency $\omega_p$. With transition matrix element $d_{eg}$, the absorption cross-section is given by $\sigma_r = k_p \alpha_i$ with $\alpha_i = 2 |d_{eg}|^2/\Gamma$, $\Gamma$ the linewidth of the $|e\rangle - |g\rangle$ transition. The atomic ensemble follows an average spatial density distribution $\varrho(r) = \langle \sum_r \delta (|r - r_i|) \rangle$ that is assumed to be nearly spherical and smooth, in particular, $\varrho(r)$ does not vary substantially on length scales other than those close to its characteristic radius $\sigma \gg \lambda_p$. We further restrict our discussion to intermediate sample with $\sigma \ll ct$, with $c$ the speed of light and $t$ the shortest time-scale of interest. The transmission of a plane-wave resonant probe beam at the exit of the atomic sample, in the $r = \{r_\perp, r_p\}$ coordinate, follows the Beer-Lambert law with transmission $T(r_\perp) = e^{-OD(r_\perp)}$. The 2D optical depth distribution is given by $OD = N \varrho (r_\perp) \sigma_r, \varrho (r_\perp) = \frac{1}{\pi} \int \varrho (r) dr_p$ the normalized column density as in the main text.

To describe both the collective dipole dynamics and its collective radiation, we regard the small atomic sample as system and free-space optical modes as reservoir. The electric-dipole interaction can be effectively described by the many-atom density matrix $\rho$, after the photon degrees of freedom being eliminated by the standard Wigner-Weiskopf procedure. Following the general approach [20] the density matrix $\rho$ obeys the master equation $\dot{\rho} = \frac{1}{i\hbar} [H_{\text{eff}}, \rho] - \rho H^\dagger_{\text{eff}} \rho + L_c |\rho|$, where $L_c$ is the ”population recycling” super-operator associated with random quantum jumps in the stochastic wavefunction picture. Here we focus on the effective Hamiltonian
$H_{\text{eff}}$ that governs the deterministic evolution of states and observables. The non-Hermitian effective Hamiltonian can be expressed as

$$H_{\text{eff}} = \sum_i H_i^a + \hat{V}_{\text{DD}, \text{eff}}$$

(B1)

with single atom Hamiltonian $H_i^a$ for atom at location $r_i$, and effective dipole-dipole interaction operator $\hat{V}_{\text{DD}, \text{eff}} = \sum_{i,j} \hat{V}_{\text{DD}}^{i,j}$ that sums over the pairwise resonant dipole interaction

$$\hat{V}_{\text{DD}}^{i,j} = \frac{\hbar k}{\varepsilon_0} d_{eg} G(\mathbf{r}_i - \mathbf{r}_j, \omega_p) d_{eg} \sigma_i^+ \sigma_j^-.$$  

(B2)

Here $\sigma_i^+ = |e_i\rangle\langle g_i|$, $\sigma_i^- = (\sigma_i^+)\dagger$ are the raising and lowering operators for the $i^{th}$ atom. The $G(\mathbf{r}, \omega_p)$ is the free-space Green’s tensor of the electric field obeying

$$\nabla \times \nabla \times G(\mathbf{r}, \omega_p) + k_0^2 G(\mathbf{r}, \omega_p) = \delta^3(\mathbf{r}).$$

(B3)

With the spin model description of the atomic dipole degrees of freedom, spontaneous emission from the atomic ensemble to free space is conveniently associated with the electric field operator:

$$\hat{E}_s(\mathbf{r}) = \frac{k_0^2}{\varepsilon_0} \sum_i N G(\mathbf{r} - \mathbf{r}_i, \omega_p) d_{eg} \sigma_i^-.$$  

(B4)

Instead of generally discussing evolution of atomic states in the $N$-spin space governed by $H_{\text{eff}}$, in the following we discuss two specific states related to this work, the timed-Dicke state $|\psi_{TD}(\mathbf{k}_p)\rangle = S^+(\mathbf{k}_p)|g_1, g_2, \ldots, g_N\rangle$, and weakly excited coherent state $|\psi(\theta_p, \mathbf{k}_p)\rangle = \prod_{i=1}^N \left(|e_i\rangle + \theta_p e^{i k_p \cdot \mathbf{r}_i} |g_i\rangle\right)$ with $\theta_p \ll 1$, keeping in mind that observables composed of collective linear operators in the two cases are intrinsically related.

We first consider the field amplitude of the spontaneously emitted photons. For $|\psi\rangle = |\psi_{TD}(\mathbf{k}_p)\rangle$, we have as in main text $\varepsilon_p(\mathbf{r}) = \langle g_1, g_2, \ldots, g_N | \hat{E}_s(\mathbf{r}) | \psi_{TD}(\mathbf{k}_p) \rangle = k_0^2 d_{eg} \frac{1}{\varepsilon_0 N} \sum_i N G(\mathbf{r} - \mathbf{r}_i, \omega_p) e^{i k_p \cdot \mathbf{r}_i}$. With the $\{\mathbf{r}_i\}$-configuration average, we have,

$$\bar{\varepsilon}_p(\mathbf{r}) = k_0^2 d_{eg} \frac{1}{\varepsilon_0 N} \int G(\mathbf{r} - \mathbf{r}', \omega_p) e^{i k_p \cdot \mathbf{r}'} d^3 \mathbf{r}'$$

(B5)

The evaluation of Eq. (B5) is aided by the fact that $\bar{\varepsilon}_p(\mathbf{r})$ is the solution to $\nabla \times \nabla \times \bar{\varepsilon}_p(\mathbf{r}) + k_0^2 \bar{\varepsilon}_p(\mathbf{r}) = \frac{k_0^2 d_{eg}}{\varepsilon_0 N} \mathbf{g}(\mathbf{r}) e^{i k_p \cdot \mathbf{r}}$ with average wave-vector $\mathbf{k}_p$ and with a slowly varying envelope (SVE). For $d_{eg}$ perpendicular to the $\mathbf{k}_p$ direction, a simplification automatically achieved by the $|\psi_{TD}(\mathbf{k}_p)\rangle$ plane-wave excitation, we end up with expression $\bar{\varepsilon}_p(\mathbf{r})$ in Eq. (1) in the main text under SVE approximation and further by ignoring transverse wave dispersion within the nearly spherical atomic sample (Raman-Nath approximation). The approximations have a fractional error on the order of $\lambda_p / \sigma$.

In the $\mathbf{r} = \{r_1, r_p\}$ coordinate Eq. (1) provides the wavefront $\bar{\varepsilon}_p(\mathbf{r}_1, 0)$ if we consider atomic sample distribution within $-\beta \sigma < r_p < 0$, with $\beta = O(1) \ll \sigma / \lambda_p$. $\bar{\varepsilon}_p(\mathbf{r}_1, 0)$ thus not only describes the average electric field amplitude at $r_p = 0$, but also the mode profile of the collective spontaneous emission in the forward direction with $r_p > 0$.

During a $\{\mathbf{r}_1\}$-specific measurement, the initial rate of photon emission into the “configuration-averaged” $\bar{\varepsilon}_p(\mathbf{r})$ mode is evaluated on the $\mathbf{r} = \{r_1, r_p = 0\}$ plane by considering the total emitted light energy into the mode divided by $\hbar \omega_p$. For the single-excitation $|\psi_{TD}(\mathbf{k}_p)\rangle$ state, the mode projection leads to $i_p^{(1)} = \frac{2 \varepsilon_0}{\hbar \omega_p} \int \bar{\varepsilon}_p(\mathbf{r}) \cdot e_p(\mathbf{r}) d^2 \mathbf{r}_1 / \int \bar{\varepsilon}_p(\mathbf{r})^2 d^2 \mathbf{r}_1$, with $e_p(\mathbf{r}) = \bar{\varepsilon}_p(\mathbf{r}) + \delta e_p$ for the particular $\{\mathbf{r}_1\}$ configuration. As the contribution from the configuration-dependent fluctuation $\delta \varepsilon_p$ scales with $1 / \sqrt{N}$, for $N \gg 1$ we have $i_p^{(1)} = \frac{2 \varepsilon_0}{\hbar \omega_p} \int \bar{\varepsilon}_p(\mathbf{r})^2 d^2 \mathbf{r}_1 \approx O(N) \Gamma / 4$ during a single measurement.

On the other hand, by defining the classical field amplitude associated with the coherent state $\varepsilon_p(\mathbf{r}) = \langle \psi(\theta_p, \mathbf{k}_p) | \hat{E}_s(\mathbf{r}) | \psi(\theta_p, \mathbf{k}_p) \rangle$, the derivation of the field amplitude for $|\psi\rangle = |\psi(\theta_p, \mathbf{k}_p)\rangle$ is very similar to the timed-Dicke states, with identical expressions for $\varepsilon_p(\mathbf{r})$ and $\bar{\varepsilon}_p(\mathbf{r})$ other than an additional factor $\sqrt{N} \theta_p$. Different from $|\psi\rangle = |\psi_{TD}\rangle$ where the phase of the photon emission is fundamentally random, for $|\psi\rangle = |\psi(\theta_p, \mathbf{k}_p)\rangle$ the collective radiation is phased according to the electric dipole coherence.

We now discuss time-dependence of collective spontaneous emission described by Eq. (2) and then by Eq. (1) in the main text. The topic is related to collective Lamb shift in a dilute atomic gas, an important and quite subtle effect well studied in previous work [30, 73, 74]. In order to apply the general theoretical predictions to this work, we explore the spin model [20] to revisit the decay part of the problem, for the quite dense and small samples here.

We consider free gas evolution with $H_{\text{eff}} = \hat{V}_{\text{DD}, \text{eff}}$ and time-dependent field amplitude $\varepsilon_p(\mathbf{r}, t) = \langle g_1, g_2, \ldots, g_N | \hat{E}_s(\mathbf{r}, t) | \psi_{TD}(\mathbf{k}_p) \rangle$, for $|\mathbf{r} - \mathbf{r}_1| \gg \lambda_p$ and with $\hat{E}_s(\mathbf{r}, t)$ evolving according to Heisenberg-Langevin equation $\dot{\hat{E}}_s = \frac{i}{\hbar} (\hat{E}_s \hat{V}_{\text{DD}, \text{eff}} - \hat{V}_{\text{DD}, \text{eff}}^\dagger \hat{E}_s) + \hat{f}$. With the Langevin force $\hat{f}$ being averaged to zero, for $|\psi\rangle = |\psi_{TD}(\mathbf{k}_p)\rangle$ we have

$$\dot{\varepsilon}_p(\mathbf{r}, t) = -i \langle g_1, g_2, \ldots, g_N | \hat{E}_s(\mathbf{r}, t) | \psi_{TD}(\mathbf{k}_p) \rangle \hat{V}_{\text{DD}, \text{eff}} |\psi_{TD}(\mathbf{k}_p)\rangle.$$  

(B6)

To evaluate Eq. (B6), we insert the orthogonal timed-Dicke basis $\{ |\psi_{TD}(\mathbf{k}_p)\rangle, |\psi_1(\mathbf{k}_p)\rangle, \ldots, |\psi_{N-1}(\mathbf{k}_p)\rangle\}$ as in ref. [30] into the equation. Here $|\psi_n(\mathbf{k}_p)\rangle = S_n^{-}(\mathbf{k}_p)|g_1, \ldots, g_N\rangle$ are single-excitation collective states with $S_n^{-}(\mathbf{k}_p) = \sum_c c_{n,c} \sigma_c^-$, $n = 1, \ldots, N - 1$ and with $c_{n,c}$ properly chosen to ensure the basis orthogonality [30]. We further define the far-field emission amplitudes associated with the $N - 1$ $|\psi_n(\mathbf{k}_p)\rangle$ states as $\varepsilon_n(\mathbf{r}, t) = \langle g_1, g_2, \ldots, g_N | \hat{E}_s(\mathbf{r}, t) | \psi_n(\mathbf{k}_p) \rangle$.
\[\langle g_1, g_2, \ldots, g_N \rangle |\hat{E}_s(r, t)\rangle |\psi_n(k_p)\rangle. \] We have,

\[\begin{align*}
\hat{V}(r, t) &= -iV_{DD}(k_p, k_p)\varepsilon_p(r, t) + Ni\n\int \varepsilon_{n}(k_p)\varepsilon_{n}(r, t), \quad (B7)
\end{align*}\]

with \(V_{DD}(k_p, k_p) = \langle \psi_{TD}(k_p)|\tilde{V}_{DD}(k_p)|\psi_{TD}(k_p)\rangle\) and similarly \(V_{DD}(n, k_p) = \langle \psi_{n}(k_p)|\tilde{V}_{DD}(k_p)|\psi_{TD}(k_p)\rangle\). The 2nd line of Eq. (B7) includes random and collective couplings between the \(k_p\) superradiant excitation and other super- and sub-radiant modes \([76]\), a fact associated with super- and sub-radiant modes \([76]\), a fact associated with the collective Lamb shifts \([30]\).

To obtain the simple expression of \(V_{DD}(k_p, k_p)\) in Eq. (B7), we rewrite the integration into the form of \(\int \varepsilon_{n}(k_p)\varepsilon_{n}(r, t)\), and the collective Lamb shifts \([30]\). To obtain the simple expression of \(V_{DD}(k_p, k_p)\) in Eq. (B7), we rewrite the integration into the form of \(\int \varepsilon_{n}(k_p)\varepsilon_{n}(r, t)\), and the collective Lamb shifts \([30]\).

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We now discuss the case of coherent state input with \(|\psi\rangle = |\psi_{\theta_p}(k_p)\rangle\). Consider \(\varepsilon_p(r_o, t) = \langle \psi_{\theta_p}(k_p)|\hat{E}_s(r_o, t)\rangle |\psi_{\theta_p}(k_p)\rangle\), again for \(r_o\) in the far field from the sample along the \(k_p\) direction. With \(\theta_p \ll 1\), we make the replacement of \(\sigma_{\theta_p}^\alpha(t)\) with \(-1\) in the Heisenberg-Langevin equation, and define \(\beta(t) = \langle \sigma_{\theta_p}^\alpha(t)\rangle\). The resulting coupled dipole model for \(\beta(t)\) is again rewritten into the form of Eq. (B7) with \(\varepsilon_p(r_o, t)\) and auxiliary \(n = 1, ..., N-1\) orthogonal mode expansion. With nearly zero initial values for the auxiliary modes, we again ignore the associated couplings for the initial dynamics so as to reach Eq. (B10). Finally we again approximate the decay rate of \(\varepsilon_p(t)\) with that for the strictly forward superradiant emission \(|\varepsilon(r_o, t)\rangle\). Combined with \(\varepsilon_p(0) = N\theta_p^2\sigma_{DD}\Gamma(4)/4\), we thus reach Eq. (4) in the main text. Finally, the discussions of Eqs. (5) in the main text is in light of intuition cast by Eq. (B7).

We remark that in all the discussions in this work, the replacement \(|d_{eg}|^2 = \hbar^2\alpha_2/2\) is general and applicable to transitions with level degeneracy. Thus we expect the conclusions for Eqs. (1)(2)(4)(5) in the main text applicable to the D2 line of 87Rb atom in this work.

2. Geometric control of collective dipole excitation, a 3-level model

We now turn to 3-level model to describe geometric control of collective dipole excitation and thus collec-
tive spontaneous emission. With the additional auxiliary state $|a\rangle$, the resonant dipole interaction in Eq. (B1) is modified as

$$\hat{V}^{ij}_{DD} = \frac{k^2}{\hbar}\hat{d}^*_{eg}G(r_i - r_j, \omega_p)\hat{d}_{eg}\sigma^+_i \sigma^-_j + \frac{k^2}{\hbar^2}\hat{d}^*_{eg}G(r_i - r_j, \omega_c)\hat{d}_{eg}\sigma^+_i \sigma^-_j,$$ \hspace{1cm} (B11)

with $\sigma^+_{i,j} = |a_i\rangle\langle g_i|$ and $\sigma^-_{i,j} = (\sigma^+_{i,j})^\dagger$. We refer the first and second line as $V^{ij}_{DD,es}$ and $V^{ij}_{DD,ec}$ respectively.

With $|e\rangle$ decoupled from the control interaction, the single atom Hamiltonian

$$H_a^\prime = \hbar \Delta |a_i\rangle\langle a_i| + \frac{\hbar}{2} (\eta(r_i)\Omega_c(t) e^{-i\varphi_c(r_i,t)} \sigma^+_i + h.c)$$ \hspace{1cm} (B12)

governs the control dynamics in the $|g\rangle - |a\rangle$ subspace. The time-dependent Rabi frequency $\eta(r_i)\Omega_c(t) = |E_c(r_i, t) \cdot \hat{d}_{ec}|/\hbar$ is driven by the control laser with a Gaussian beam intensity profile, with $\Omega_c(t)$ the peak value and $\eta(r_i)$ a position dependent factor. The control phase $\varphi_c(r_i, t)$ is written in the rotating frame detuned from the $|g\rangle - |a\rangle$ transition by $\Delta$. The goal is to design $\Omega_c(t)$, $\varphi_c(r_i, t)$ so that any quantum state of many atoms governed by the master equation associated with Eq. (B1) undergoes state-dependent phase-patterning as in Eq. (3) in the main text, and therefore $U^{i}_{c}\varphi_c = 1 + e^{i\varphi_c(r_i,t)}|g_i\rangle\langle g_i|$ for all the N atoms.

To implement the geometric phase patterning, we consider multiple pulse control as in this work with $\Omega_c(t)$ and $\varphi_c(r_i, t)$ split into $n$ smooth sections arriving at $t = t_n$. For a single pulse $n$, we consider $\varphi_{c,n}(r_i, t) = \delta_{c,n}(t-t_n)$. By redefining $|a_i\rangle$ with $e^{-i\int^t \sum_{c,n} \delta_{c,n}(t-t_n)dt'} \sigma^+_i + h.c)$ phase factor, Eq. (B12) can be rewritten in the new rotating frame as

$$H_a^{\prime} = \frac{\hbar}{2} \sum_{c,n} (\Delta + \delta_{c,n}(t-t_n)) (\hat{1}_{g,a} + \sigma^-_{c,i}) + \frac{\hbar}{2} \sum_{c,n} (\Delta - \delta_{c,n}(t-t_n)) (\hat{1}_{g,a} + \sigma^-_{c,i}) + h.c),$$ \hspace{1cm} (B13)

with $\hat{1}_{g,a} = |a_i\rangle\langle a_i| + |g_i\rangle\langle g_i|$ and $\sigma^-_{c,i} = |a_i\rangle\langle g_i| - |g_i\rangle\langle a_i|$.

Exploring the SU(2) symmetry in Eq. (B13), it is straightforward to achieve $U^{i}_{c}\varphi_c$, in absence of dipole-dipole interaction or spontaneous emission, by successively applying two nearly identical $|g\rangle - |a\rangle$ inversion pulses with $\delta_{c,1} = \delta_{c,2}$, $\Omega_{c,1} = \Omega_{c,2}$ but with different optical phase $\varphi_1, \varphi_2(r_i)$. In particular, we consider the state evolution $|\psi(t)\rangle = |g_i\rangle\psi_1(\tau_c + \tau_d)$ subjected to $n = 1, 2$ population inversion pulses, each with a $\tau_c$ duration, and with $t_1 = 0$ and $t_2 = \tau_d$. The “return amplitude” of cyclic evolution $f_g = |g_i\rangle\psi_1(\tau_c + \tau_d) = |f_g\rangle e^{-i\varphi_c} \sigma^+_i \sigma^-_j$ is characterized by $\varphi_c(r_i) = \varphi_1 + \varphi_2$ including dynamic $\varphi_D$ and geometric $\varphi_G$ phases. For an ideal pair of population inversion pulses, $|f_g\rangle = 1$, and the SU(2) symmetry suggests

$$\varphi_G(r_i) = \pi + \varphi_1(r_i) - \varphi_2(r_i)$$ \hspace{1cm} (B14)

determined by the optical phase difference between the otherwise nearly identical pulse pair. The $\varphi_G$ is visualized on the Bloch sphere (Fig. 1 in main text) as half the solid angle spanned by the cyclic state trajectory. With the 2-level symmetry, the dynamic phase $\varphi_D = (\int_0^{\tau_d} dt + \int_{\tau_d}^{\tau_c} dt)\psi_1(t)H_a^{\prime}\psi_1(t))$ for the perfect inversions can be expressed as:

$$\varphi_D(r_i) = \int_0^{\tau_c} \varphi_1(t)dt + \int_0^{\tau_d} \varphi_2(t)dt,$$ \hspace{1cm} (B15)

with $h_1$ given by the $n = 1, 2$ terms in the summation of Eq. (B13) excluding the $\varphi_1, \varphi_2$ part respectively.

To arrive at both Eqs. (B14)(B15), we assume $\eta_1 \approx \eta_2$ and $|\psi_1(\tau_d < t < 2\tau_d)\rangle$ approximately follows $|\psi_1(t < t < \tau_d)\rangle$ on the Bloch sphere up to a rotation. Any spatially-dependent $\varphi_{D,a}(r_i)$ is nullified if the two inversion pulses are with identical strength so that $\eta_1 = \eta_2$. The additional, spatially independent phase $\varphi_{D,d}(r_i)$ is nullified by the $n = 1, 2$ terms in the summation of Eq. (B13) excluding the $\varphi_1, \varphi_2$ part respectively.

To achieve $\Delta$ and $\eta(r_i)$ independent population inversion, the simplest choice is a quasi-adiabatic pulse. With $\Omega_c = \Omega_0 \sin(\pi t/\tau_c)$ and $\delta_{c} = -\delta_{c} \cos(\pi t/\tau_c)$, stability of near unity inversion efficiency against $\Delta$ and $\eta$ has been studied in detail in the context of nuclear magnetic resonance [78], molecular spectroscopy [79], and matter-wave accelerations [80][71]. Efficient and error-resilient inversion is achievable with $(\Omega_0, \delta_0)$ close in magnitude and for $\int \Omega_cdt$ beyond $3\pi$, as in this work.

We now perturbatively estimate the influence of resonant-dipole interaction by Eq. (B11) to the geometric phase patterning of collective dipole excitation. For simplicity we only consider the timed Dicke state $|\psi(0)\rangle = |\psi_{TD}(k_p)\rangle$. With the retro-reflected traveling wave pulses and $\varphi_G(r_i) = \pi + 2\delta_{c}$, a perfect single atom operation leads to $|\psi(\tau_c + \tau_d)\rangle = U_c(\varphi_G)|\psi_{TD}(k_p)\rangle = e^{i\delta_{c} - 2\delta_{c}k_p}\psi_{TD}(k_p - 2k)\rangle$ in absence of $V^{ij}_{DD}$. Error associated with the resonant dipole interaction can be estimated with either incoherent phase error $\delta \varphi_i = \sqrt{\delta \varphi_i^2}$, or the collective phase error $\delta \varphi_N = \sqrt{N\delta \varphi_i^2}$, with

$$\delta \varphi_i^2 = \sum_i \left( \sum_j \int_0^{\tau_c + \tau_d} |\psi(t)|^2 V^{ij}_{DD,es} \psi(t)|dt|^2 \right),$$

$$\delta \varphi_N^2 = \sum_i \left( \sum_j \int_0^{\tau_c + \tau_d} |\psi(t)|^2 V^{ij}_{DD,es} \psi(t)|dt|^2 \right)$$ \hspace{1cm} (B16)

for contribution from the $V^{ij}_{DD,es}$ and $V^{ij}_{DD,ec}$ interaction respectively. The error is evaluated with the unperturbed $|\psi(t)\rangle$ evolving according to the single-body $H_{cl}, H_{c2}$ control. The perturbative treatment is valid for $\delta \varphi_i \ll 1$, which is generally achievable with increased $(\Omega_0, \delta_0)$ and reduced $\tau_c + \tau_d$ control interval.

We consider the “worst case scenario” where control errors due to all the pairwise interactions add up coherently to perturb the collective control dynamics, with overall error characterized by $\delta \varphi_N$. We also consider the shortest possible duration $\tau_c + \tau_d = 2\tau_c$. In
light of the fact that the collective error is associated with collective interaction, it is straightforward to have \( \delta \varphi_{N,c} \sim \tau_c \sqrt{(\delta_{N,eg})^2 + (\Gamma_{N,eg})^2/4} \), that scales with the largest collective Lamb shift \( \delta_{N,eg} \) and decay rate \( \Gamma_{N,eg} \) of the singly excited gas [20, 30]. With the \( |g\rangle - |a\rangle \) population inversions and phase imprinting, \( \delta_{N,eg} \) and \( \Gamma_{N,eg} \) reduce substantially during the control. The symbol \( \langle ... \rangle \) averages over the instantaneous values, leading to at least a factor of 50% reduction to the collective part of \( \delta \varphi_{N,c} \).

Resonant dipole interaction on the strongly driven \( |g\rangle - |a\rangle \) transition is much stronger than on the weakly excited \( |g\rangle - |e\rangle \) transition. Accordingly, \( \delta \varphi_{N,a} \sim N\tau_c \sqrt{(\delta_{N,eg})^2 + (\Gamma_{N,eg})^2/4} \) can be much larger. However, in contrast to \( \delta \varphi_{N,c} \), it is not appropriate to directly associate \( \delta \varphi_{N,a} \) with control error since the collective radiation addresses the same \( |g\rangle - |a\rangle \) transition as the “very strong” control \( \Omega_c \). In fact, during the control the collective dipole radiation amounts to absorbing and reshaping the \( \Omega_c (r, t) e^{-i\varphi_{r,t}} \) control pulse, and the reverse effects can be largely suppressed by the quasiadiabatic technique insensitive to the pulse shape for population inversions [30]. The collective radiation thus impacts the phase patterning operation as a collective dynamic phase shift according to Eq. (B15), which can be quite uniform across the atomic sample and do not contribute to the actual collective dipole control error. With a concrete study of the open system coherent control for future work [31], we conclude this section by suggesting that the control error due to resonant dipole interactions depends on details of atomic position arrangements, and could be bounded by \( \delta \varphi_{M} \sim \max(\delta \varphi_{N,c}, \delta \varphi_{i,a}) \) with careful choice of \( \Omega_c, \varphi_G \) to avoid substantially distortion of the shape-optimized control pulses by the atoms, or simply by implementing the control instead on excited state transitions [33] in which case \( \delta \varphi_A \) becomes much less important.

3. Single atom model for the experiment

To model the experiments with a dilute gas of moderate OD and further with \( \Gamma_{D2}(\tau_c + \tau_d) \approx 0.1 \) in this work, we simplify Eq. (B1) by absorbing the imaginary parts of \( V_{DDI}^{ij} \) into \( H_0 \) and then ignore the rest of resonant dipole interactions. This leads to effective Hamiltonian

\[
H_{eff} = \sum_{j=1}^{N} \left( H_{a} - i \frac{\Gamma_{D2}}{2} |e_j\rangle \langle e_j| - i \frac{\Gamma_{D1}}{2} |a_j\rangle \langle a_j| \right). \tag{B17}
\]

Here \( H_0^{ij} \) is according to Eq. (B12). By ignoring atom-atom interaction during the \( \tau_c + \tau_d \) interval, the numerical modeling of the control dynamics is expected to be accurate at \( \Gamma_{D2}(\tau_c + \tau_d)/(\overline{OD})/4 \approx 3\% \) level for evaluating the collective dipoles, according to discussions in the last sections. Here we expect \( \overline{OD} < 0.5 \overline{OD}_p \) due to the \( |g\rangle - |a\rangle \) population inversion and inhomogenous dynamic phase writing that substantially reduce the optical depth seen by the \( k_p \) or \( k_s \) radiation during the control of the nearly spherical sample.

With the simplified Hamiltonian in Eq. (B17) that ignores atom-atom interaction, we are now free to choose \( r_i \), for notation convenience, in particular, we change the basis for single atom wavefunction into \( k- \) space, with \( |g, k\rangle = \frac{1}{\sqrt{N}} \sum_i e^{i k r_i} |g_i\rangle \) and similarly for \( |e, k\rangle \) and \( |a, k\rangle \), by adjusting \( r_i \) to ensure the orthogonality of the \( k- \) state basis of interest. In addition, we expand the level structure to that of the \( ^{87}\text{Rb} \) D1 and D2 line, and use \( \{ g, e, a \} \) as indices to label the \( \{ 5S_{1/2}, 5P_{3/2}, 5P_{1/2} \} \) hyperfine levels respectively. We end up with \( H_{eff} = \sum H_{eff}^{(s)} \) composed of single-atom Hamiltonian in \( k- \) space, with

\[
H_{eff}^{(s)} = H_p + H_c + H_{e,c}.
\]

\[
H_p = \sum_{g,k} \Delta_g |g, k\rangle \langle g, k| + \sum_{e,k} \Delta_e |e, k\rangle \langle e, k| + \sum_{a,k} \Delta_a |a, k\rangle \langle a, k| + \sum_{e,g,a,k} \left( \frac{1}{2} \Omega_p \langle t + t_p \rangle c_{eg}^{h.c.} |e, k + k_p\rangle \langle g, k| + h.c. \right),
\]

\[
H_c = \sum_{g,a,k} \delta_c \langle t - t_1 \rangle |a, k\rangle \langle a, k| + \sum_{g,a,k} \left( \frac{1}{2} \eta_1 \Omega_c \langle t - t_1 \rangle c_{ag}^{h.c.} |a, k + k_c\rangle \langle g, k| + h.c. \right),
\]

\[
H_{e,c} = \sum_{g,a,k} \delta_e \langle t - t_2 \rangle |a, k\rangle \langle a, k| + \sum_{g,a,k} \left( \frac{1}{2} \eta_2 \Omega_e \langle t - t_2 \rangle c_{ag}^{h.c.} |a, k - k_e\rangle \langle g, k| + h.c. \right). \tag{B18}
\]

Here to be specific we consider a probe excitation during \( -\tau_p < t < 0 \), followed by two control pulses \( H_{c1}, H_{c2} \) during \( t_1 = \Delta t_1 + t_\tau \) and \( t_2 = \Delta t_1 + \tau_d \) as in Fig. [1] in the main text to make the \( U_c \) phase-patterning control. The \( c_{ag}^{\pm} \) are combinations of Clebsch-Gorden coefficients to characterize the D1 and D2 transitions driven by the \( x- \) and \( y- \) polarized control and probe lasers respectively. We also introduce \( \eta_{1,2} \) factors similar to those in Eq. (B12) to account for laser intensity inhomogeneities.

Clearly, Equation (B18) can also be interpreted as being written in the space-time domain with quantized atomic wavefunction, without kinetic energy terms. Indeed, the atomic motion within the sub-nanosecond control in this work can be ignored, and we adapt this wavefunction interpretation when using the same equations to calculate both dipole control and optical acceleration.

To write down the single-atom master equation, we introduce six “effective” collapse operators

\[
\hat{C}_{D1}^{ij} = \sum_{c,g,k} \sqrt{\Gamma_{D1} c_{eg}^{h.c.}} |g, k\rangle \langle c, k + k_p|
\]

\[
\hat{C}_{D2}^{ij} = \sum_{c,g,k} \sqrt{\Gamma_{D2} c_{eg}^{h.c.}} |g, k\rangle \langle c, k + k_p| \tag{B19}
\]

with “\( j \)” running through “\( x \), “\( y \)”, and “\( z \)” polarizations. The collapse operators are associated with quantum jumps and spontaneous emission. We effectively set the recoil \( k- \)shifts in simple ways to minimize the calculation complexity, without affecting the \( D_2 \) dipole coherence and the \( D_1 \) optical force under study.

We are now able to write down the master equation
for the single-atom density matrix $\rho^{(s)}$ as

$$\dot{\rho}^{(s)}(t) = \frac{i}{\hbar}(H_{\text{eff}}^{(s)} - \rho^{(s)}H_{\text{eff}}^{(s)})^\dagger + \sum_j (C_{D1}^{(j)}\rho^{(s)}C_{D1}^{(j)} + C_{D2}^{(j)}\dot{\rho}C_{D2}^{(j)}). \tag{B20}$$

With $\rho^{(s)}(t)$ it is straightforward to calculate the interaction-free evolution of many-atom density matrix $\rho(t) = (\rho^{(s)}(t))^{\otimes N}$ and to evaluate collective observables $\langle \hat{O} \rangle = \text{tr}(\rho(t)\hat{O})$. It is important to point out that the simple single-body method is incapable of describing initially entangled states such as timed-Dicke states. However, as in quantum optics, as long as the observables to be evaluated are only composed of linear dipole operators, their dynamics during the interaction-free evolution should be captured by dynamics of weakly excited coherent states.

We further simplify Eq. (B18) by restricting the momentum basis according to situation of our experiments. In particular, we set the initial condition $\rho^{(s)} = \frac{1}{2}\sum_{|g,k}\langle g,k|q(g,k)|g,k\rangle$ running through the $|F = 2, m_F\rangle$ Zeeman sublevels. The k states are coupled to $k + k_p$ and $k \pm nk_c$ via the probe and control interactions. By ignoring atomic motion, we only consider a single k-class as illustrated by the “momentum lattices” in Fig. 5 which also highlight the structure of the couplings according to Eqs. (B18) (B19) (B20). We restrict the accessible momentum states with $|n| < 6$ for the numerical calculations. The truncation is validated by numerically monitoring the high-n states and by verifying the consistent results with larger n-cutoffs.

With experimental imperfections encoded in parameters like $\eta_{1,2}$ in Eq. (B18), we refer to the numerically evaluated single-atom density matrix according to Eq. (B20) as $\rho^{(s)}(t)$. For comparison, the perfect geometric phase transfer is achieved by replacing the evolution by $H_{C1} + H_{C2}$ in Eq. (B18) with instantaneous $U_c = 1 - \sum_{g} |g,k\rangle \langle g,k| + \sum_{g} |g,k+2k_c\rangle \langle g,k|$, leading to “perfectly controlled” density matrix $\rho^{(s)}_0(t)$ for $t > 0$.

With $\rho^{(s)}(t)$ we evaluate the dipole coherence $\langle d(k_s) \rangle = \text{tr}(\rho^{(s)}(t)d(k_s))$ for the weakly and coherently excited gas, with coherence operator $d(k_s) = \sum_{g,c} c_{g,c}^{\dagger}g|e,k + k_p\rangle\langle e,k + k_p|$. The collective coherence in the main text is related to $\langle d(k_s) \rangle$ as $\langle S^-(k_s) \rangle \propto N\langle d(k_s) \rangle$, and furthermore we approximate $\langle S^+(k_s)S^-(k_s) \rangle \approx \langle S^-(k_s) \rangle^2$ for $N \gg 1$. We thus in addition evaluate $i_{s,\eta}(t) = N^2\langle \text{tr}(\rho^{(s)}(t)d(k_s)) \rangle$ and similarly $i_{s,0}(t) = N^2\langle \text{tr}(\rho^{(s)}(t)d(k_s)) \rangle$ for redirected superradiance under perfect control.

4. The $f_d$ and $f_a$ estimation

We relate the experimental observable $i_s(t)$ with ensemble-averaged $i_{s,\eta}(t)$ as $\langle i_{s,\eta}(t) \rangle$, and calculate collective dipole control efficiency as $f_d = \langle i_{s,\eta}(t)\rangle_{\eta}/i_{s,0}(0)$. The ensemble average of emission intensity, instead of field amplitude, is in light of fact that we experimentally collect $i_s(t)$ with a multi-mode fiber, and the signal $i_s(t)$ is insensitive to slight distortion of the $E_r$-mode profile by the dynamic phase writing according to Eq. (B13) due to the imbalanced $\eta_{1,2}$.

The simulation of optical acceleration by the D1 control pulses follows the same Eqs. (B18) (B20), but without the probe excitation and with atomic levels restricted to the D1 line only. We evaluate the momentum transfer as $\Delta P_n = h\sum_{g,n}2n|g,k+2nk_c\rangle \langle \rho^{(s)}(t)|g,k+2nk_c\rangle + \sum_{g,n}(2n+1)|g,k+(2n+1)k_c\rangle \langle \rho^{(s)}(t)|g,k+(2n+1)k_c\rangle$ for $t = \tau_c + \tau_d$. We then compare the ensemble-averaged acceleration efficiency $f_a = \langle \Delta P \rangle_{\eta}/(2hk_c)$ with the experimental measurements.

The $\eta_{1,2}$ average in both calculations is according to spatial distribution of control laser beam intensity profile seen by the atomic sample. As the final results are quite insensitive to distribution details, we assume both the laser beam and the atomic sample with gaussian profiles, with waists $w = 13 \mu m$ and $\sigma = 7 \mu m$ by fitting the imaging measurements and with optics simulations. We adjust the retro-reflected beam waist $w_r$ and the intensity factor $\eta_2 \propto 1/w_r$ accordingly in the simulation, together with an overall intensity calibration factor $\kappa$ multiplied to the $s$ parameter from the beat-note measurements (Appendix A). The ensemble averaged $f_a$ is compared with experimentally measured $\Delta P/2hk_c$, and we adjust $\kappa, w_r$ to globally match the single-atom simulation with all the measurement results for optical acceleration as in Fig. 4. We then estimate both $f_a, f_d$ as discussed in Sec. II.D.

5. Superradiance suppression and recall

To further simulate the experimental sequence (Fig. 1), it is straightforward to add the 2nd phase-patterning control $U_c(\phi_G)$ and the “recall” control $U_c^{-1}(\phi_G)$, by adding corresponding pulsed interactions to the Hamiltonian Eq. (B18). We then calculate $\langle |d(k)^2| \rangle = \langle |\text{tr}(\rho^{(s)}(t)d(k_s))| \rangle_{\eta}$ associated with collective dipole ex-

FIG. 8. “Momentum lattice” structure for probe excitation and $U_c$ control simulations according to Eqs. (B18) (B19) (B20). Dash arrows represent the “effective” quantum jump operations associated with Eq. (B19). The double-sided arrows represent the coherent laser couplings. The coherence between the wavily underlined lattice sites $|e,k + k_p\rangle$ and $|g,k + 2k_c\rangle$ is associated with the redirected superradiant emission.
citation with $k = \{k_p, k_s = k_p - 2k_c, k'_s = k_p - 4k_c\}$ for the forward, redirected, and “subradiantly stored” collective radiation respectively. In according with the experimental settings here, we again follow the “intensity average” instead of “amplitude average” for ensemble average of collective dipoles.

At the experimental side, implementation of the 2nd $U_c$ operation is straightforward by applying a 2nd shaped pulse to the atoms, which is automatically followed by the retro-reflected pulse after the $\tau_d$ optical delay. The implementation of $U_c^{-1}$ operation is however limited by the $\tau_d = 1.36$ ns delay line. With modified implementation of $U_c^{-1}$ operation for a future paper, in this work we resorted to a Kapitza-Dirac diffraction [21], with interaction written in $k$–space as

$$H_{KD} = \sum_{a,k} \Delta_{KD}(t - t_p)|a, k\rangle \langle a, k| + \sum_{g,a,k} \frac{1}{2} \Omega_{KD}(t - t_r) c_{ag}^\dagger(a, k - k_c)|g, k| + h.c. + \sum_{g,a,k} \frac{1}{2} \Omega_{KD}(t - \tau_d) c_{ag}^\dagger(a, k + k_c)|g, k| + h.c. \quad (B21)$$

Here $t_r = \Delta t_1 + \Delta t_2 + 3(\tau_1 + \tau_d)$ is the proper delay according to Fig. 4 for the approximately sine-shaped recall pulse $\Omega_{KD}(t) = \Omega_{KD, M} \sin(\pi t / \tau_{KD})$. With the duration $\tau_{KD} = 10$ ns $> \tau_d = 1.36$ ns, the optical delay by the retro-reflecting mirror can be ignored and the light field forms a standingwave with amplitude following $\Omega_{KD}$ in time. The detuning of the standingwave to the D1 line is set as a constant $\Delta_{KD}$ during the same $\tau_{KD}$ duration. To interpret Eq. (B21) with the momentum lattice picture in Fig. 3, the standingwave couples $k$ to $k \pm k_c$ that can be coherently continued across the lattices to large $n$. Correspondingly, in the simulation we make a higher cutoff of $|n| < 20$ for reliable simulations.

With the simulation parameters optimally matching the experiments, we not only reproduce features of experimental observable $i_s(t) \propto \langle |d(k_p)|^2 \rangle$, but also unveil time-dependent dynamics for the un-monitored forward emission $\langle |d(k_p)|^2 \rangle$ and the “subradiantly stored” or the superradiance-free excitation $\langle |d(k'_s)|^2 \rangle$. Typical results are given in Fig. 9.

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