Precise spinor matterwave control with nanosecond adiabatic spin-dependent kicks

Liyang Qiu, Lingjing Ji, Yizun He, Jiangyong Hu, Yuzhuo Wang, and Saijun Wu

1Department of Physics, State Key Laboratory of Surface Physics and Key Laboratory of Micro and Nano Photonic Structures (Ministry of Education), Fudan University, Shanghai 200433, China.

(Dated: today)

Significant aspects of advanced quantum technology today rely on rapid Raman control of atomic hyperfine matterwaves. Unfortunately, efficient Raman excitations are usually accompanied by uncompensated dynamic phases and coherent spin-leakages, preventing accurate and repetitive transfer of recoil momentum to large samples. We provide systematic study to demonstrate that the limitations can be substantially overcame by dynamically programming an adiabatic pulse sequence. Experimentally, counter-propagating frequency-chirped pulses are programmed on an optical delay line to parallelly drive five \( \Delta m = 0 \) hyperfine Raman transitions of \(^{87}\text{Rb}\) atoms for spin-dependent kick (SDK) within \( \tau = 40 \) nanoseconds, with an \( f_{\text{SDK}} \approx 97.6\% \) inferred fidelity. Aided by numerical modeling, we demonstrate that by alternating the chirps of successive pulses in a balanced fashion, accumulation of non-adiabatic errors including the spin-leakages can be managed, while the dynamic phases can be robustly cancelled. Operating on a phase-stable delay line, the method supports precise, fast, and flexible control of spinor matterwave with efficient Raman excitations.

I. INTRODUCTION

Precise control of 2-level systems is instrumental to modern quantum technology. In atomic physics, such controllable two-level systems are naturally defined on a pair of long-lived atomic internal states and are often referred to as atomic spins. When the atomic spins are controlled by optical Raman transitions, quantized photon recoil momentum can be transferred to the center-of-mass motion during a spin-flip [1–3]. By laser cooling [4], the atomic motion can be sufficiently slowed that the spin-dependent momentum transfers driven by rapid optical pulses are effectively instantaneous "spin-dependent kicks" (SDK) [5, 6] with an accuracy insensitive to initial atomic velocity. SDKs are therefore a class of broadband spinor matterwave control techniques with an achievable accuracy similar to those for 2-level internal atomic spin controls [7–9]. Beyond traditional applications such as to enhance the enclosed area of light pulse atom interferometers [3, 6, 10], SDKs emerge as an important technique to control spin-motion entanglement and to improve the scalability of ion-based quantum information processing [5, 11–13].

For coherent control of spinor matterwave, it is essential to operate SDK at high enough speed so as to suppress effects from low-frequency perturbations and to compose multiple operations within a limited duration. Furthermore, except for working with microscopically confined samples [5, 8, 13], high quality optical control needs to be highly resilient to intensity errors associated with illumination inhomogeneity [14, 15]. To meet the fidelity requirements by the next generation quantum technology [16–21], particularly for large samples, high-speed implementation of error-resilient composite techniques [22–24] are likely required. However, the operation speed of SDK are practically limited by intricate requirements for precise Raman control, including the following two categories.

The first type of SDK speed constraints are associated with the suppression of spin leakage and dynamic phase. To avoid spin leakage into excited states causing spontaneous emission, the Raman excitation bandwidth has to be much smaller than the single-photon detuning \( \Delta_e \) (Fig. 1(a)). However, since the Raman coupling strength is inversely proportional to \( \Delta_e \), a small \( \Delta_e \) is required to support efficient control with limited laser power. Furthermore, keeping \( \Delta_e \) smaller than the ground-state hyperfine splitting \( \omega_{\text{hfs},g} \) is important for the Raman scheme to accurately suppress the dynamic phase associated with differential Stark shifts (Fig. 1(b)) [25]. These \( \Delta_e \) constraints strongly limit the speed and fidelity of the Raman control. In addition, as to be clarified further, for Raman control with moderate \( \Delta_e \) in alkaline atoms, the excited state hyperfine interaction leads to tensorial \( m \)-changing transition [26] to invalidate the 2-level spin picture in the first place. The standard method to avoid the problem is to lift the Zeeman degeneracy with a strong enough quantization field [27], which in turn limits the speed of the Raman control.

The second type of SDK speed constraints are associated with directionality of SDK. As in Fig. 1, when counter-propagating \( \mathbf{E}_{1,2} \) pulses are applied to drive a Raman transition, to ensure the preference for the atom to absorb a photon from the \( \mathbf{E}_1 \) field followed by a stimulated emission into the \( \mathbf{E}_2 \) field, certain mechanism is needed to prevent the time-reversed Raman process from occurring. For hyperfine spin control of alkaline-like atoms, the directionality is ultimately supported by the ground-state hyperfine splitting \( \omega_{\text{hfs},g} \) [5]. Practically, however, when the Raman interferometry is operated in the retro-reflection geometry with established advantages [28–33], the SDK speed is limited by the typically moderate 2-photon frequency differences introduced by additional frequency modulations [3, 28] or Doppler.
FIG. 1. Schematic of Raman SDK on a hyperfine manifold, with the $^{85}\text{Rb}$ D1 line in this work as the example. (a) Cross-linearly polarized, counter-propagating $E_1, E_2$ resonantly drive the $\Delta m = 0$ and $\Delta m = \pm 2$ Raman transitions. When the single-photon detuning $\Delta_e$ is much larger than the excited state hyperfine splitting $\omega_{hfs,e}$, the $\Delta m = \pm 2$ couplings through the $F' = 2 \leftrightarrow F'' = 3$ intermediate states largely cancel each other, leading to decoupled $\{|a_m\rangle, |b_m\rangle\}$ sub-spin systems. (b) The reduced level diagrams for the weakly coupled $\{|a_m\rangle, |b_m\rangle\}$ systems ($m = \pm 1$). The bare levels are relatively shifted and Raman coupled. The effective Hamiltonian parameters $\Omega^{(m)}, \Omega^{(m+2)}_R$ for Eqs. (1)(2)(3) are marked. The differential Stark shift $\delta_0$ is largely responsible for the SDK dynamic phase (Eq. (5)). Similarly coupled sub-spin systems composed of $\{|a_m\rangle, |b_m\rangle\}$ with $m = 0, \pm 2$ are not shown.

FIG. 2. (a) The schematic setup. Frequency-chirped optical waveforms (illustrated with the real part of the complex envelop function $\mathcal{E}_{1,2}$, sharing a same color-coding as those for $E_{1,2}$ in Fig. 1(a)) are programmed by an optical arbitrary waveform generator (OAWG) and sent through a single mode fiber to the delay line. An $E_1$ pulse collides with a retro-reflected $E_2$ pulse at the atomic sample to drive the $|a_m\rangle \leftrightarrow |b_m\rangle$ Raman transition while imparting $\pm \hbar k_R$ kicks to atoms reaching $|a_m\rangle$ and $|b_m\rangle$ respectively. Next, after a delay time $\tau_d$, the $E_1$ pulse is retro-reflected to meet $E'_2$ at the same location to further boost the $|a_m\rangle, |b_m\rangle$ separation. QWP: quarter-wave plate. (b) The timing sequence for the “balanced chirp-alternating scheme” (Eq. (11)). The two-photon detunings $\delta_R$ are plotted vs time. The adiabatic controls with “up-chirp” and “down-chirp” 2-photon detuning $\delta_R$ are referred to as $U_u$ and $U_d$ respectively. Here the amplitudes for the retro-reflected pulses are reduced by a $\kappa$ factor. Typical Bloch sphere dynamics of an $\{a_m, b_m\}$ spinor ($m = 0$) are given in (c) at various laser intensities parametrized by the Raman pulse area $A_R$. The weak $\Delta m = \pm 2$ couplings (Fig. 1(b), Eq. (1)) are ignored.

The purpose of this work is to improve the speed and precision of spinor matterwave control in presence of the above mentioned technical barriers. In particular, we study Raman adiabatic SDK [6, 10] within nanoseconds in a retro-reflection optical setup [32, 34], for alkaline atoms with Zeeman degeneracy prone to coherent spin leakage. As schematically summarized in Fig. 1 (level diagram) and Fig. 2 (experimental implementation), counter-propagating frequency-chirped pulses are programmed on the optical delay line to adiabatically drive the $F_b = 2 \leftrightarrow F_a = 3$ Raman transitions of $^{85}\text{Rb}$ through the D1 line, while imparting $\pm \hbar k_R$ momentum “kicks” to atoms. Here $k_R = k_2 - k_1$ is the difference of $k$-vectors between the pulse pair. The adiabatic Raman transfer technique [6, 10, 35] (Fig. 2(b)(c)) provides the intensity-error resilience for the focused laser to address...
a mesoscopic sample. To enforce the directionality in the standard retro-reflection geometry [32, 34], the regular method of resolving the 2-photon frequency differences [3, 28, 29] are abandoned. Instead, we exploit the fact that the nanosecond pulses are short enough to be spatially separated (Fig. 2(a)), allowing controllable collision of specific Raman pulses within the atomic sample, at specific instances, to drive the Raman transitions.

Experimentally, equipped with a wideband optical arbitrary waveform generation system (OAWG) [36], we drive adiabatic SDKs within tens of nanoseconds, the fastest realization to date [6, 10], by shaping counter-propagating pulses (Fig. 2) with merely tens of milliwatts laser power. The spin population and momentum transfer are measured and compared with full-level numerical simulations, with which we infer an SDK fidelity of $f_{\text{SDK}} \approx 97.6(3)\%$. The $\delta f \sim 2.5\%$ infidelity, as unveiled by the full-level numerical simulation, is primarily limited by spontaneous emission and the $\Delta$ unveiled by the full-level numerical simulation, is primarily limited by spontaneous emission and the $\Delta m = \pm 2$ leakage among Zeeman sublevels within the ground state hyperfine manifold (Fig. 1(b)) at the moderate $\Delta_e = 2\pi \times 10$ GHz detuning.

Aided by numerical modeling, we further demonstrate that beyond the incoherent momentum and population transfer, high-fidelity spinor matterwave “phase gates” capable of coherently transferring many recoil momenta can be synthesized by nanosecond adiabatic SDKs on the delay line. This is despite the coherent spin leakage and intensity-dependent diffraction phase broadening [6, 25, 37, 38] by individual “kicks”, at the moderate $\Delta_e$ [8, 39]. In particular, we demonstrate that a tailored adiabatic sequence with alternating up and down 2-photon chirps (Fig. 2(b)(c)) suppresses the coherent accumulation of spin leakages across the nearly degenerate hyperfine levels dressed by the cross-linearly polarized pulses (Fig. 1(b)). Furthermore, as to be clarified further, the time-reversal symmetry of the chirp-balanced scheme ensures robust cancellation of the laser intensity-dependent diffraction phases, with $\Delta_e > \omega_{\text{hfs},g}$ unbounded by traditional choices [25, 40], even in presence of non-perfect retro-reflection (Fig. 2(b)) [32].

In the following the main part of the paper is structured into three sections. In Sec. II, we set up a light-atom interaction framework where multi-level couplings are treated as perturbations to the effective 2-level Raman interaction within the ground state hyperfine manifold. Key notations and quantities for the experimental and numerical studies are defined. In Sec. III we demonstrate the flexible matterwave control by programming counter-propagating pulses on an optical delay line with OAWG [36]. Constrained by experimental resources, the characterization of the adiabatic SDK technique is limited to the inference of the SDK fidelity with atomic velocity and hyperfine population measurements. However, in Sec. IV we demonstrate efficient suppression of coherent spin-leakage accumulation and intensity-dependent dynamic phases by the chirp-alternating scheme, and provide numerical evidence that the chirp-balanced SDK scheme support coherently control of multi-spinor matterwave with large momentum transfer, even with a moderate laser power in the 10 mW range as in this work.

II. THEORETICAL MODEL

A. Spin-dependent kicks on a hyperfine manifold

We consider the Fig. 1 (a) D1 transition of an alkali-like atom. The Zeeman-degenerate $a, b$ and $e$ hyperfine states with total angular momentum $F_{a,b}$ (single-valued) and $F'_e$ (multi-valued for both hyperfine levels) are labeled as $|c_m\rangle \equiv |c, F, m\rangle$ with magnetic quantum number $m \in [-F_e, F_e]$. Here, with the nuclear spin $I > 1/2$, there are $2F_e + 1$ Zeeman sublevels for each manifold $c \in \{a, b\}$, and similarly for the $e$ manifold. The $|a_m\rangle \leftrightarrow |b_n\rangle$ hyperfine Raman transition is driven by pairs of counter-propagating laser pulses, $E_{1,2} = \epsilon_{1,2} \hat{E}_{1,2} e^{i(k_e x - \omega_e t)} + c.c.$ with shaped slowly-varying amplitudes $\hat{E}_{1,2}(\mathbf{r}, t)$ and Raman-resonant carrier frequency difference $\nu_2 - \nu_1 = \omega_{\text{hfs},g}$.

Although motion of the multi-level atom in the pulsed $E_{1,2}$ fields is quite complicated (Appendix A), the internal state dynamics can be substantially simplified when the laser polarization $E_{1,2}$ are chosen to be cross-linear (Fig. 2(a)) [30, 41] and the single-photon detuning $\Delta_e$ is much larger than the excited state hyperfine splitting $\omega_{\text{hfs},c}$. In this case, the destructive interference of Raman couplings through the intermediate $|c_\ell\rangle$ levels prevent efficient $\Delta m = \pm 2$ transitions (The rule in Fig. 1(a) is that efficient Raman couplings are only composed by pairs of dipole couplings drawn with a same type of solid or dashed lines.) [26]. The residual “leaking” Raman Rabi frequency (Fig. 1(b)) scales as

$$\Omega_{\text{R}} = \mathcal{O} \left( \frac{\omega_{\text{hfs},c}}{\Delta_e} \right) \Omega_{\text{R}},$$

with $\Omega_{\text{R}} = \Omega_{a}^{(1)} \Omega_{b}^{(2)} / 2\Delta_e$, $\Omega_{a}^{(2)}(b) = \frac{\epsilon_{1}(a)}{\hbar} \frac{1}{\sqrt{3}} \langle |J_{y}| |d||J_{x}| \rangle$ [42].

Therefore, in the large $\Delta_e$ limit, the ground-state manifold is decomposed into those within $2F_e + 1$ copies of pseudo spin-1/2 sub-spaces $\{\{a_m\}, |b_n\rangle\}$ labeled by magnetic quantum number $m$ (See Fig. 1(b) for the $m = \pm 1$ examples). For each $m$-spin, the Rabi frequency driven by the Raman pulse can be written as

$$\Omega_{\text{R}}^{(m)} = \chi^{(m)} \Omega_{\text{R}}.$$

The close-to-unity factor $\chi^{(m)}$ is determined by the associated Clebsh-Gorden coefficients, normalized at $\chi^{(0)} = 1$ and decreases slowly with $|m|$. The Hamiltonian for the $m$-spin is given by

$$H_{0}^{(m)}(\mathbf{r}, t) = \hbar \left( \frac{\delta_0}{2} \sigma_{z}^{(m)} + \frac{\Omega_{\text{R}}^{(m)}}{2} e^{i k_{R} \cdot r} \sigma_{+}^{(m)} + h.c. \right).$$

Here the detuning $\delta_0$ is the $(m$-independent) differential Stark shift between the two hyperfine ground states that
can be nullified at suitable $\Delta;/\omega_{\text{hf},g}$ and $\Omega_{a,b}$ combinations [25]. The Pauli matrices $\{\sigma_x^{(m)}, \sigma_y^{(m)}, \sigma_z^{(m)}\}$ are defined by $\sigma_+^{(m)} \equiv \sigma_{a_{m}b_{m}}^{(m)}$ [44]. For notation convenience in the following, we further define projection operators $1^{(m)} \equiv \sigma_{a_{m}a_{m}}^{(m)} + \sigma_{b_{m}b_{m}}^{(m)}$ and $1^{(m)} = 1 - 1^{(m)}$ respectively.

With the major part of the resonant Raman interaction identified, the D1 multi-level dynamics (Fig. 1(a)) governed by the effective light-atom interaction Hamiltonian, as detailed in Appendix A, can be written as

$$H_{\text{eff}}(r, t) = \sum_{m=-F_k}^{F_k} H^{(m)}_{\text{eff}}(r, t) + V'(r, t) \tag{4}$$

to conveniently describe the Raman-dressed ground state dynamics. Apart from the $\Delta m = \pm 2$ couplings specified by Eq. (1), the $V'$ term includes non-Hermitian and non-adiabatic $|e\rangle$ couplings, the $m$-sensitive light shifts [26], and the “counter-rotating” couplings involving detuned Raman excitations (Eq. (A3)).

It is important to note that the atomic position $r$ parameter in Eq. (4) can be regarded as a quantum mechanical operator acting on the external atomic wavefunction. The Eq. (4) Hamiltonian can therefore propagate the spinor matterwave together with the kinetic $\hat{P}^2/2M$ operator, see Appendix B for implementation ($\hat{P} = i\hbar\nabla$ and $M$ the atomic mass.) During nanosecond intervals, however, the matterwave dispersion for the laser-cooled atoms is negligibly small. Opportunity is therefore open for designing broadband matterwave controls for their iterative applications. In particular, a spin-dependent kick is a transfer of photon momentum to atoms accompanied by a spin-flip [5, 6]. We refer an ideal SDK as

$$U_K(k_R) = \prod_{m=-F_k}^{F_k} \left( e^{i\sigma_+^{(m)}k_R r} - \text{h.c.} + 1^{(m)} \right). \tag{5}$$

The $\varphi^+$ is a diffraction phase offset [45] which is generally Zeeman state $m$- and laser intensity $|\mathcal{E}|^2$-dependent.

In the following our goal is to construct perfect $U_K(k_R)$ operation as in Eq. (5) from the full Hamiltonian in Eq. (4), and furthermore to design multiple SDKs with suppressed $m$ and $|\mathcal{E}|^2$ dependence. For the purpose, we first define a fidelity $f_{\text{SDK}}$ to qualify the implementation.

### B. $f_{\text{SDK}}$ and $\varepsilon_{\text{leak}}$ for non-ideal SDK

To qualify the Fig. 2 SDK implementation modelled by Eq. (4) Hamiltonian, we refer the imperfect realization of SDK as $\tilde{U}(k_R; \eta)$ with $\eta$ to generally represent relevant Hamiltonian parameters. We define an average fidelity [46] over the whole atomic ensemble for a single SDK acting on the $\{|b_m\}, \{|a_m\}\}$ states as

$$f_{\text{SDK}} = \left\langle \left| \left\langle c_m U_K^\dagger(k_R) \tilde{U}(k_R; \eta) c_m \right| \right\rangle \right\rangle_{\eta,c_m}^{2}. \tag{6}$$

Here the $\tilde{U}(k_R; \eta) c_m$ is compared to $U_{\text{ref}}(k_R) c_m$ across the atomic sample with an $\langle \ldots \rangle_z$ average, effectively performing a Fourier transform. An ensemble average of the mode-squared fidelity is then performed with $\langle \ldots \rangle_\eta$ over the Hamiltonian parameters of interest. Assuming light intensity hardly varies along $z$ over the wavelength-scale distance of interest (Fig. 2(a)), the SDK fidelity defined this way becomes insensitive to the diffraction phase $\varphi^+$ and therefore provides a convenient measure for the quality of controlling incoherent observables, such as for the recoil momentum and hyperfine population transfer to be experimentally measured next. The $\langle \ldots \rangle_{c_m}$ instead performs average over the $2(2F_k+1)$ initial states of interest with $m = -F_k, ..., F_k$ and $c = a, b$.

To quantify the leakage of atomic state out of each spin sub-space $\{|a_m\}, |b_m\rangle$ by $V'(r, t)$, we define an average spin leakage probability by the non-ideal SDK as

$$\varepsilon_{\text{leak}} = 1 - \left\langle \left\langle c_m \tilde{U}(k_R; \eta) 1^{(m)} \tilde{U}(k_R; \eta) c_m \right| \right\rangle_{\eta,c_m}. \tag{7}$$

Since spin leakage leads to inefficient control, we generally expect $f_{\text{SDK}} \leq 1 - \varepsilon_{\text{leak}}$. The leakage probability $\varepsilon_{\text{leak}}$ defined by Eq. (7) includes contributions from $\Delta m = \pm 2$ transition as well as those due to spontaneous emission, as $\varepsilon_{\text{leak}} = \varepsilon_{\text{sp}} + \varepsilon_{\text{sm}}$. Here the spontaneous emission probability $\varepsilon_{\text{sp}}$ during the SDK control is obtained by evaluating $\tilde{U}(k_R; \eta)$ with the stochastic wavefunction method [47, 48] (Appendix B). For the $\Delta m = \pm 2$ leakage, we find $\varepsilon_{\text{sm}} \propto \omega_{\text{hf},e}^2 / \Delta_z^2$, as by Eq. (1), and is thus suppressible at large $\Delta_z$ similar to the suppression of spontaneous emission. However, one should note that unlike spontaneous emission which simply leads to decoherence, the $\Delta m = \pm 2$ leakage is a coherent process. A sequence of leakage driven by multiple SDKs may interfere constructively to amplify the effect, even if $\varepsilon_{\text{sm}}$ for a single SDK is negligibly small.

### C. Adiabatic SDK

To achieve a uniformly high SDK fidelity across an intensity-varying sample volume, particularly for $m-$spin on a hyperfine manifold with different $f_R^{(m)}$ (Fig. 1, Eq. (2)), a standard technique is to exploit the geometric robustness of 2-level system by inducing an adiabatic rapid passage (Fig. 2(a)) [6, 10, 35, 49, 50]. For a particular $\{|a_m\}, |b_m\rangle$ sub-spin, if we ignore $V'$ in Eq. (4), then the Raman adiabatic passage can easily be generated by the $H^{(m)}_{\text{eff}}$ Hamiltonian. In particular, we parametrize the Rabi frequency of the two pulses as $\Omega_{a(b)}^{(1)} = C_{a(b)} e^{i\phi_{a(b)}^{(1)}}$, and specifically consider the time-dependent phase difference and the amplitude profile as $\Delta \phi(t) = \phi_0 \sin(\pi t/\tau_c)$, $C_{a(b)}(t) = C_{a(b)}(0) \sin(\pi t/\tau_c)$ respectively for $t \in [0, \tau_c]$ (Fig. 3(b) inset). With a large enough 2-photon sweep frequency $\delta_{\text{swp}} = \pi \phi_0 / \tau_c$, $|\delta_{\text{swp}}| \gg \delta_0$ to let the 2-photon detuning $\delta_R = \Delta \phi$ cover the differential Stark shift $\delta_0$ (Fig. 2(b)), a strong enough Raman
Rabi amplitude $C_R^{(0)} = C_b^{(0)} C_a^{(0)}/2\Delta_c = 2\chi^{(m)} A_R/\tau_c$ with a peak Raman pulse area $A_R \gg 1$, and matched magnitudes between $C_R^{(0)}$ and $\delta_{\text{swp}}$, SDK can be generated in a quasi-adiabatic manner [51], i.e., with atomic state $|\varphi_m(t)\rangle = c_a(0) e^{i\varphi_a(t)} |\varphi_m(t)\rangle + c_b(0) e^{i\varphi_b(t)} |\varphi_m(t)\rangle$ following the adiabatic basis $\{|a_m(t)\rangle, |b_m(t)\rangle\}$ which are simply the eigenstates of the instantaneous Hamiltonian. Population inversion are thus driven quasi-adiabatically during $0 < t < \tau_c$ with the efficiency insensitive to the laser intensity, detuning, and their slow deviations from the specific time-dependent forms. Putting back the $r$-dependence, the diffraction phase accompanying the population inversion, $\hat{\varphi}^+(r) = \hat{\varphi}^+ + k_R \cdot r$, is evaluated as $\hat{\varphi}^+(r) = \varphi_a(\tau_c) - \varphi_b(\tau_c)$ which includes not only a geometric phase $\varphi_G = \pi/2 + k_R \cdot r$ [52, 53], but also a dynamic phase $\varphi_{D,m} \propto \int_0^{\tau_c} (\langle \hat{a}_m | H_0^{(m)} | \hat{a}_m \rangle - \langle \hat{b}_m | H_0^{(m)} | \hat{b}_m \rangle) d\tau$ sensitive to the control parameters $\Omega^{(2)}_{ab}$ characterized by the laser intensity profiles.

### D. Dynamic phase cancellation

Clearly, for different $m$-spins and in presence of intensity $|\mathcal{E}|^2$ inhomogeneities, the diffraction phase $\varphi^+$ (Eq. (5)) printing into the matterwave is not controlled. Therefore, even the perfect $U_K^{(2N)}(k_R)$ cannot control the spinor matterwave coherently by itself (Fig. 1(a)). It is worth noting that the dynamic phase for the adiabatic SDK survives at vanishing $\delta_0$ [25, 53]. Therefore, coherent matterwave control with adiabatic SDK requires certain dynamic phase cancellation in general.

The dynamic phases for perfect SDKs can be cancelled in pairs. To avoid trivial operations during the pairing, the 2-photon wavevector $k_R$ can be inverted [3, 6, 10], leading to

$$U_K^{(2N)}(k_R) = U_k(-k_R) U_k(k_R) \cdots U_k(-k_R) U_k(k_R)$$

$$= \prod_{m=-F_b}^{F_b} \left(-1\right)^N e^{i 2N k_R \cdot r_0(t_m) + \hat{\mathcal{I}}^{(m)}}.$$  

Within each $m$-spin space span by $\{|a_m\rangle, |b_m\rangle\}$, the $U_K^{(2N)}(k_R)$ acts as a position-dependent phase gate to pattern the two components of arbitrary spinor matterwave with $\pm (2Nk_R \cdot r)$ phases, i.e., to coherently transfer opposite photon recoil momenta to the spin components.

Practically, however, swapping the $k$-vectors of $\mathcal{E}_{1,2}$ can affect other pulse parameters [28, 29, 32, 54]. Here, the $k$-vector swapping is achieved on the delay line with retro-reflection (Fig. 2) by programming the carrier frequencies $\nu_1 \leftrightarrow \nu_2$ for the delayed pulses. The imperfect reflection with $k < 1$ generally leads to unbalanced diffraction phases [34] $\hat{\varphi}^+$ associated with $\hat{U}(\pm k_R)$. For the spinor matterwave control, we quantify the impact of dynamic phase by comparing the phase of imperfectly controlled $\hat{U}(2N)|c_m\rangle$ with the ideal $U_K^{(2N)}|c_m\rangle (c = a, b)$,

$$\varphi_{D,m}^{(2N)} = \text{arg}(\langle a_m | r_k^{(2N)}(k_R) U(k_R | \eta) a_m \rangle) - \text{arg}(\langle b_m | r_k^{(2N)}(k_R) U(k_R | \eta) b_m \rangle).$$  

### E. Chirp-alternating adiabatic SDK schemes

A central goal of this work is to synthesize perfect $U_K^{(2N)}(k_R)$ spinor matterwave phase gates with $2N$ imperfect $\hat{U}(\pm k_R)$, even in presence of the unbalanced dynamic phase $\varphi_{D,m}^{(2N)}$ and the $\Delta m = \pm 2$ leakage. For the purpose, we refer the experimentally realized non-ideal adiabatic SDKs with equal amount of positive ($\delta_{\text{swp}} > 0$) and negative ($\delta_{\text{swp}} < 0$) 2-photon sweeps as $\hat{U}_u(\pm k_R)$ and $\hat{U}_d(\pm k_R)$ respectively (Fig. 2(b)). The associated control Hamiltonians are $H_u(r, t)$ and $H_d(r, t)$. In Sec. IV C we show that a chirp-alternating sequence

$$\hat{U}_{ud}^{(2N)}(k_R) = \hat{U}_u(-k_R) \hat{U}_d(k_R) \cdots \hat{U}_u(-k_R) \hat{U}_d(k_R),$$

fairly efficiently suppress the $\Delta m = \pm 2$ leakage. Next, as detailed in Sec. IV C, Sec. IV D, by combining the $U_{ud}^{(2N)}$ and $U_{du}^{(2N)}$ to form a balanced chirp-alternating sequence,

$$\hat{U}_{uddu}^{(2N)}(k_R) = \hat{U}_{ud}^{(2N)}(k_R) \hat{U}_{du}^{(2N)}(k_R),$$

the dynamic phases by $\hat{U}_{du}^{(2N)}(k_R)$ and $\hat{U}_{ud}^{(2N)}(k_R)$ robustly cancel each other. The Eq. (11) scheme can therefore faithfully realize $U_K^{(2N)}(k_R)$ phase gate.

### III. EXPERIMENTAL IMPLEMENTATION

#### A. Nanosecond SDK on a delay line

The adiabatic SDK is implemented on the $^{85}\text{Rb} 5S_{1/2} - 5P_{1/2}$ D1 line as depicted in Fig. 1, with counter-propagating chirp pulses programmed by a broadband optical waveform generator [36] on an optical delay line [15]. The cross-linear polarization is realized by double-passing the light beam with a quarter waveplate before the end mirror (Fig. 1(b)) which converts the incident $e_x$ polarization to $e_y$. With the OAWG output peak power limited to $P_{\text{max}} \approx 20$ mW, the incident control pulse $\mathcal{E}_{1,2}$ is focused to a waist radius of $w \approx 13 \mu m$ to reach a peak Rabi frequency of $\Omega_{a(b)} \approx 2 \pi \times 2$ GHz. The imperfect retro-reflection with $\kappa \approx 0.7$ ($r = \kappa^2 \approx 50\%$) reflectivity, primarily limited by increased focal beam size due to wavefront distortion, leads to decreased $\Omega_{b(a)} = \kappa \Omega_{a(b)}$ for the reflected pulses seen by the atomic sample. We set the single-photon detuning to be $\Delta_e = 2 \pi \times 10$ GHz to achieve a peak Raman Rabi frequency of $C_R(0) \approx 2 \pi \times 300$ MHz estimated at the center of the Gaussian $\mathcal{E}_{1,2}$ beams. An $\tau_d = 140.37$ ns optical
delay is introduced by the $L \approx 20$ m folded delay line, which is long enough to spatially resolve the counter-propagating nanosecond pulses. To form the counter-propagating $E_{1,2}$ pulse pair, we pre-program $E_{1,2}(t)$ and $E_{2,1}(t - \tau_d)$, with a relative delay matching the optical delay line, to ensure the pulse pair with proper carrier frequency $\nu_{1,2}$ meeting head-on-head in the atomic cloud. To continue multiple SDKs, additional, individually shaped pulses with alternating $\nu_{1,2}$ can be applied, as in Fig. 2(b), with a $T_{\text{rep}} = \tau_d$ periodicity.

Importantly, to periodically generate multiple Raman SDKs with the delay line, every shaped pulse contribute twice to the SDK sequence, except for a “pre-pulse” and “post-pulse” which meet the atomic sample alone without any counter-propagating pulses to help driving the Raman transition (Fig. 2(b)). To properly shape the Raman coupling $\Omega_R(t) \propto E_1^*(t) E_2(t)$ (Eq. (2)), therefore, one needs to program the incident $E_{2(1)}^*$ pulse according to the retro-reflected $E_{2(1)}$ pulse. To clear out such pulse-history dependence in a long sequence, the retro-reflection cycles can be interrupted a few times by increasing the interpulse delays beyond $\tau_d$. The interruptions would lead to more pairs of “pre-pulses” and “post-pulses”. However, these extra pulses impact negligibly the atomic hyperfine population and momentum transfer. Their impacts to interferometric applications are discussed in Sec. V.

Here, if our goal is to alternate Raman chirps $\delta_{1,2}$ as in Fig. 2(b) and Eq. (11), then the frequency-sweeping amplitudes for every optical pulse needs to be increased by an additional $\delta_{\text{swp}}$ (Sec. II C). As a result, in the balanced chirp-alternating scheme (Eq. (11)) the first 2N pulses becomes more and more chirped, before a reversal of the process to rewind back the rate. High optical chirping rates affect the $V'$-couplings (Eq. (4)). In our experiments, the single-photon detuning $\Delta_\nu = 2\pi \times 10$ GHz is much larger than $\delta_{\text{swp}}$, and we have numerically confirmed that the increasingly chirped waveforms do not significantly affect the Raman dynamics in the $U_{\text{udud}}^{(4N)}$ scheme up to $N = 6$. Nevertheless, to avoid systematic errors associated with the digital AWG pulse shaping [36], the chirp-rate accumulation is interrupted in this work by separating $U_{\text{udud}}^{(4N)}$ into N sets of 4-pulse sequences, as described above.

B. Optimizing Adiabatic SDK

We prepare $N_A \sim 10^5 ^{85}$Rb atoms in a compressed optical dipole trap at a temperature of $T \sim 200$ µK [36]. The atomic sample is optically pumped into the $F = 2, |m = 3\rangle$ hyperfine states, elongated along $z$, with a characteristic radius of $\sigma \approx 7$ µm in the x-y plane (Fig. 3(a)). Immediately after the atoms released from the trap, multiple SDKs programmed on the optical delay line with alternating $\pm k_R$, $k_R = 2k_0 e_z$ are applied to transfer photon momentum by repetitively inverting the atomic population between $F = 2, |m = 3\rangle$ and the $F = 3, |m = 2\rangle$ hyperfine states. Here $k_0 = 2\pi/\lambda$ is the wavenumber of the D1 line SDK pulses at $\lambda = 795$ nm.

We use a double-imaging technique to characterize the performance of the adiabatic SDKs, by simultaneous measuring the resulting spin-dependent momentum transfer and population inversion (Appendix D). Specifically, immediately after the last of $n$ SDK pulses, a probe pulse resonant to the D2 line $F = 3 - F' = 4$ hyperfine transition is applied for $\tau_p = 20$ µs to record the spatial distribution of atoms in state $|a\rangle$, in the $x - z$ plane, with calibrated absorption imaging [15]. Next, after a $\tau_{\text{tof}} = 160$ µs free-flight time, the 2nd $\tau_p = 20$ µs resonant probe pulse is applied to image all the atoms. For the purpose, during the time of flight an additional 50 µs pulse along $e_x$, resonant to $F = 2 - F' = 3$ transition, repumps the $|b\rangle$ atoms to $|a\rangle$ for the 2nd imaging. By comparing atom number $N_a$ in state $|a\rangle$ and the total atom number $N_A = N_a + N_b$, inferred from the first and second images respectively, the probability of atoms ending up in $|a\rangle$ can be measured as a function of the number of SDKs $n$ as $\rho_{aa,n} = N_a/(N_a + N_b)$. In addition, by fitting both images to locate the center-of-mass vertical positions $z_{1,2}$, the atomic velocity $v_n = \delta z/\tau_{\text{tof}}$ can be retrieved to estimate the photon momentum transfer $p_n = Mv_n$ in unit of $\hbar k_R$.

Typical $v_n$ measurement results are given in Fig. 3(b). Here, for atoms prepared in $|b\rangle$ states subjected to an $n = 25$ SDK sequence starting with $U_a(k_R)U_a(-k_R)$ (a

FIG. 3. (a) SDK momentum transfer measurement with a double imaging method: For atomic sample subjecting $n$ SDKs ($n = 25$ here) starting with $U_a(k_R)U_a(-k_R)$, a sequence of two absorption images are taken at $t_1 = 10$ µs and $t_2 = t_1 + \tau_{\text{tof}}$, separated by $\tau_{\text{tof}} = 160$ µs time-of-flight and with a hyperfine repumping pulse along $x$ applied in between (see main text). The center-of-mass position shift $\delta z$ is retrieved with a typical ±2 µm accuracy by fitting the optical depth (OD). The recoil velocity $v_n = \delta z/\tau_{\text{tof}}$ is then estimated during typical adiabatic SDK parameter scan as in (b). Here, with $n = 25$, $\tau_p = 60$ ns SDK pulses applied, $v_n$ is optimized as a function of sweep frequency $\delta_{\text{swp}}$, and peak laser intensity parametrized by an estimated peak Raman pulse area $A_R = C_R \tau_p/2$. Blue, green and orange lines correspond to estimated peak $A_R$ of approximately $6\pi$, $9\pi$, $12\pi$ respectively. The corresponding peak Rabi frequencies $C_R$ are marked with arrows on the top along the $\delta_{\text{swp}}$ axis. The error bars give statistical uncertainties of $v_n$ in repeated measurements.
2N = 24 double-SDK followed by an additional kick to drive the final Raman transition), the atomic population is largely in |a⟩ while vn is unidirectional along e_z. We optimize vn by varying the peak Raman coupling amplitude CR(0) and sweep frequency δswp of the adiabatic SDK pulses at fixed τc. As in Fig. 3(b), for a fixed peak Raman pulse area A_R, we generally find δswp to be optimized for efficient photon momentum transfer when it matches CR(0) (See the arrow markers in Fig. 3(b)). However, unlike 2-level transfer [51] where an increased pulse area always leads to improved adiabaticity and population inversion robustness, here we find the peak A_R ≈ 9π reaches optimal to ensure the resilience of adiabatic SDK against the up to 50% intensity variation in the setup. Larger A_R is accompanied by slow decrease of vn, due to increased probability of spontaneous emission. Here, for τc = 60 ns, we need to attenuate CR(0) ≈ 2π × 150 MHz to keep the peak A_R ≈ 9π. By using the full CR(0) ≈ 2π × 200 MHz available in this work, we are able to reduce τc to 40 nanoseconds while maintaining nearly identical momentum transfer efficiency at δswp = 2π × 150 MHz.

C. Inference of fSDK

With the optimal δswp = 2π × 150 MHz and peak A_R ≈ 9π at τc = 40 ns, we now apply n = 1–25 SDKs to characterize the momentum transfer p_n = Mvn and normalized population ρaa,n as a function of kicking number n. Typical results are given in Fig. 4. Here the momentum change p_n along e_z is again unidirectional along z as in Figs. 4(a,i). The direction is conveniently reversed by programming ˜U_u(−kr) first in the ±kr alternating sequence, resulting in acceleration of atoms along −e_z instead as in Figs. 4(a,ii). In contrast, the hyperfine population ρaa,n is suppressed and revived after an even and odd number of SDKs respectively, as demonstrated in Fig. 4(b). In addition, we program a balanced chirp-alternating sequence n = 4N that combines ˜U_u(±kr) with ˜U_d(±kr) as G_{udda}(±kr) according to Eq. (11), a sequence which will be detailed in Sec. IV for interferometric applications, with p_n and ρaa,n measurement results also given in Fig. 4 to demonstrate similar momentum and population transfer efficiency.

We estimate fSDK by comparing the p_n and ρaa,n measurements as in Fig. 4 with precise numerical modeling detailed in Appendix B, taking into account the finite laser beam sizes and imperfect reflection with reflectivity r = |κ|^2. The comparison is facilitated by fitting both the measurement and simulating data according to a phenomenological model (Appendix C), which assumes that errors between successive SDKs are uncorrelated and are solely parametrized by fR, a hyperfine Raman population transfer efficiency. The model predicts exponentially reduced increments |p_{n+1} − p_n|/ℏκ_eff = |ρaa,n+1 − ρaa,n| = fR(2fR−1)^n for fR ≈ 1 by each SDK. From measurement data in Fig. 4(a,b), fR ≈ 98.8% can be estimated in both ˜U_u(kr) and ˜U_u(−kr) kicks (Fig. 4(a)), slightly less than spontaneous-emission-limited fR ≈ 99.2% predicted by numerical simulation of the experiments, assuming perfect reflection with κ = 1.

We simply attribute the slightly reduced fR from the theoretical value to imperfect retro-reflection (Appendix E). In particular, we numerically find fR reduces with fSDK when ε1,2 are unbalanced in amplitudes, so that both spin leakage and spontaneous emission are increasingly likely to occur. Taking into account the independently measured R ≈ 0.5 in this work and with numerically matched fR, ε_leak ≈ 2.5% with ε_{Δm} ≈ 0.5% and ε_sp ≈ 2% can be estimated respectively. We therefore infer from the combined analysis an fSDK = 97.6(3)% for the adiabatic SDK in this experiment, slightly less than the spontaneous-emission-limited population transfer.
\( f_{\text{SDK}} \approx 98\% \) for the nearly perfect adiabatic SDK. The numbers can be improved further by increasing \( \Delta \) to suppress spontaneous emission as well as the \( \Delta m \)-leakage. It is important to note that while the imperfect reflection only moderately reduce \( f_{\text{SDK}} \) here, the resulting imperfection of \( \mathbf{k}_R \leftrightarrow -\mathbf{k}_R \) swapping in successive \( \hat{U}_u(\pm \mathbf{k}_R) \) control can greatly compromise in the cancellation of dynamic phase \( \varphi_B \) in double SDK (Eq. (8)), a topic to be detailed in Sec. IV D.

The adiabatic SDK demonstrated in this experimental section is the fastest realization to neutral atoms to date \([6, 10]\). By equipping a more powerful laser, the SDK time can be further reduced to sub-nanosecond level \([5]\). By equipping a more powerful laser, the SDK time can be further reduced to sub-nanosecond level \([5]\), only limited by the ground state hyperfine splitting \( \omega_{\text{hfs}} \). At such high speed an optical delay line of less than one meter is long enough to resolve counter-propagating pulses for conveniently performing \( \pm \mathbf{k}_R \) kicks with suppressed systematics.

### IV. COHERENT CONTROL OF SPINOR MATTERWAVE

In the previous section, we experimentally characterize nano-second adiabatic SDK by measuring the transfer of photon momentum and hyperfine population by a SDK sequence. A natural question to ask is whether it is possible to exploit the technique for coherent spinor matterwave control. As discussed in Sec. II D, for ideal SDKs the double SDK sequence (Eq. (8)) can be constructed to perform position-dependent phase gates. However, as to be illustrated in the following, the operation degrades rapidly with kick number \( N \) in presence of coherent spin leakage among the \( 2F_1+1 \) \( \{ |a_m\rangle, |b_m\rangle \} \) sub-spins (Sec. IV B), or when the non-perfect \( \mathbf{k}_R \leftrightarrow -\mathbf{k}_R \) swapping introduces additional dynamic phases (Sec. IV D).

Nevertheless, in this section we demonstrate that the \( \Delta m \)-leakage and dynamic phases can be efficiently suppressed by the balanced chirp-alternating SDK sequence \( \hat{U}^{(2N)}_{(2k)}(\mathbf{k}_R) \) introduced in Sec. II E (Eq. (11)) for faithful implementation of the \( \hat{U}_{(2k)}^{(2N)}(\mathbf{k}_R) \) phase gate by Eq. (8) to finely enable high-efficiency large momentum transfer \([3]\). We further numerically demonstrate the utility of the tailored adiabatic SDK sequence by simulating an area-enhanced atom interferometry sequence, using the experimental parameters both within and beyond this experimental work.

We notice control of spin leakage in quasi-two-level systems is an important topic in quantum control theory \([55–57]\). Previous studies on the topic typically involve a Morris–Shore transformation of the interaction matrix to decompose the multi-level dynamics \([58]\). However, as being schematically summarized in Fig. 1(b), the spin leakages are coherently driven through multiple paths with multiple Raman couplings to preclude a straightforward transformation, nor a direct application of the associated leakage-suppression techniques \([55–57]\).

#### A. Average gate fidelity

To evaluate an imperfect double-SDK sequence \( \hat{U}^{(2N)}_{(2k)}(\mathbf{k}_R; \eta) \) as a quantum gate, we define an average fidelity for its performance on arbitrary spinor matterwave states of interest, e.g., spatially within the focal laser beam (Fig. 2(a)) and internally span the \( \{ |a_m\rangle, |b_m\rangle \} \) sub-spin space. The average fidelity is written as \([46]\)

\[
\mathcal{F}^{(2N)}_m = \left| \langle \psi_{m,j} | \hat{U}^{(2N)}_{(2k)}(\mathbf{k}_R; \eta) | \psi_{m,j} \rangle \right|^2.
\]

Similar to Eq. (6), here \( \eta \) represents the Hamiltonian parameter in Eq. (4) including the atomic position \( \mathbf{r} \) of interest. For internal states the evaluation samples \( | \psi_{m,j} \rangle \) as the six eigenstates of \( \sigma_z^{(m)} \) listing as \( \{ | \psi_{m,j} \rangle \} = \{ |a_m\rangle, |b_m\rangle, (|a_m\rangle \pm |b_m\rangle)/\sqrt{2}, (|b_m\rangle \pm i|a_m\rangle)/\sqrt{2} \} \). With external motion ignored, \( \mathcal{F}^{(2N)}_m \approx 1 \) ensures that the effectively instantaneous \( \hat{U}^{(2N)}_{(2k)}(\mathbf{k}_R; \eta) \) faithfully apply the phase gate to arbitrary spinor matterwave states near the beam focus, with certain laser intensity of interest.

For the convenience of related discussions, we define an average \( \Delta m \)-leakage probability, similar to Eq. (7), as

\[
\varepsilon_{m,\Delta m} = 1 - \langle \langle \psi^{(n)}_{m,j} | 1^{(m)} \rangle \langle \psi^{(n)}_{m,j} \rangle \rangle_{\eta,j}.
\]

where \( | \psi^{(n)}_{m,j} \rangle = \hat{U}^{(n)}_{(k)}(\mathbf{k}_R; \eta) | \psi_{m,j} \rangle \) is defined as the final atomic state after the imperfect control. To exclude spontaneous emission, we simply renormalize to have \( \langle \psi^{(n)}_{m,j} | \psi^{(n)}_{m,j} \rangle = 1 \) before the evaluation of the \( \Delta m \)-leakage. Similar to Eq. (13), here the \( \Delta m \)-leakage probability is also evaluated for a specific \( m \)-subsystem of interest. Similar to Eqs. (6)(7), here we expect \( \mathcal{F}^{(n)}_m \leq 1 - \varepsilon_{m,\Delta m} \) since any spin leakage results in gate infidelity.

#### B. Simple double-SDK: spin leakage

We define the non-ideal realization of the double-SDK sequence with \( \hat{U}_u(\pm \mathbf{k}_R) \) or \( \hat{U}_d(\pm \mathbf{k}_R) \) the same way as in Eq. (8) for the ideal case, which are referred to as \( \hat{U}^{(2N)}_{(2k)}(\mathbf{k}_R) \) and \( \hat{U}^{(2N)}_{(2d)}(\mathbf{k}_R) \) respectively in the following. We numerically investigate the \( \Delta m \)-leakage probability \( \varepsilon_{m,\Delta m} \) and the gate fidelity \( \mathcal{F}^{(n)}_m \), for an atom initialized in an \( \{ |a_m\rangle, |b_m\rangle \} \) subsystem being subjected to non-ideal double-SDK sequence \( \hat{U}^{(n)}_{(u)}(\mathbf{k}_R) \). The Hamiltonian parameters in the simulation follow the experimental setup described in Sec. III for the \( ^{85}\text{Rb} \) D1 line scheme (in particular \( \delta_{\text{rep}} = 2\pi \times 150 \) MHz), except here the reflective coefficient is set as \( \kappa = 1 \) for simplicity, and \( \Gamma = 0 \) to focus on the coherent control dynamics. To elucidate the role of laser intensity for the coherent control, we repeat the simulation while scanning the laser intensities in proportion, parametrized by the Raman pulse area \( A_R \) in the following. Typical results for \( ^{85}\text{Rb} \) are shown in the left panel of Fig. 5(a-d,i) and Fig. 6(a-c,i).
presented in Fig. 5(a,i). Here, for \( n \) ending up in a different sub-spin as a function of kicking number \( N \) with increased controls with \( n \) with increased intensity dependent manner. The fold is accompanied by partial or full returns in a laser-

FIG. 5. Numerical simulation of \( \Delta m \)-leakage dynamics. The parameters for the simulations are chosen according to the experimental Sec. III. (a): \( \varepsilon_{\Delta m} = \varepsilon_{m,\Delta m}^{(n)} \) as a function of pulse number \( n \) for the \( \tilde{U}_{uu}^{(n)}(k_R) \) (left) and \( \tilde{U}_{ud}^{(n)}(k_R) \) (right) controls with \( \mathcal{A}_R = 9\pi \). (b-d): \( \varepsilon_{\Delta m} = \varepsilon_{m,\Delta m}^{(4N)} \) for \( m = 0,1,2 \) sub-spin as a function of pulse area \( \mathcal{A}_R \) for the \( \tilde{U}_{uu}^{(4N)}(k_R) \) (left) and \( \tilde{U}_{ud}^{(4N)}(k_R) \) (right) controls. The corresponding \( f_{\text{SDK}} \)s are displayed on the top. The 2-photon detuning profiles are illustrated above the figures.

We first discuss the average \( \Delta m \)-leakage probability \( \varepsilon_{m,\Delta m}^{(n)} \) as a function of kick number \( n \) for typical \( \mathcal{A}_R = 9\pi \) presented in Fig. 5(a,i). Here, for \( n = 1 \), the tiny \( \varepsilon_{m,\Delta m}^{(n)} \) for both \( m = 0,1 \) are close to the single kick leakage \( \varepsilon_{\Delta m} \approx 0.5\% \) as being inferred experimentally. However, with increased \( n \), \( \varepsilon_{m,\Delta m}^{(n)} \) increases rapidly to approach unity for merely \( n \sim 20 \), i.e., atoms starting from any of the subsystems \( \{|a_m|,|b_m|\} \) has a substantial probability of ending up in a different \( m \)-subspace. We also note that with increased \( n \) the leakage within the hyperfine manifold is accompanied by partial or full returns in a laser-intensity dependent manner.

To investigate the laser intensity dependence, the \( \Delta m \)-leakage \( \varepsilon_{m,\Delta m}^{(2N)} \) is further plotted in Figs. 5(b-d,i) as a function of both kicking number \( n = 4N \) and pulse area \( \mathcal{A}_R \), for \( m = 0,1,2 \) subsystems respectively (The choice of \( n = 4N \) is for comparison with the balanced chrip-alternating sequence to be discussed shortly). We see that over a broad range of pulse area \( \mathcal{A}_R \), even with the near unity \( f_{\text{SDK}} \) (Fig. 5(b,i), top), the spin-leakage probability \( \varepsilon_{m,\Delta m}^{(4N)} \) increases rapidly with \( 4N \) in oscillatory fashions. There is hardly any continuous region of laser intensity with \( \varepsilon_{m,\Delta m}^{(4N)} \approx 0.1 \).

It is important to note that although the strong \( \Delta m \)-leakages hardly affect the photon momentum and hyperfine population transfer (Sec. III), they do limit the gate fidelity for faithful spinor matterwave control with the multiple SDKs. The impact of spin leakage to average gate fidelity \( F_m^{(4N)} \) is demonstrated by comparing Figs. 5(b-d,i) with Figs. 6(a-c,i), where we see the gate infidelity \( 1 - F_m^{(4N)} \) closely follows \( \varepsilon_{m,\Delta m}^{(4N)} \) to hardly reach 0.1 over most laser intensities, except when the laser intensity is too low to adiabatically drive the Raman transition at all (with \( \mathcal{A}_R < 3\pi \) here) where we instead find \( F_m^{(4N)} \approx 0.5 \), as expected.

Clearly, the coherent accumulation of \( \Delta m \)-leakage error as in Figs. 5(a-d,i) needs to be efficiently suppressed before the adiabatic SDK sequence can be exploited for coherent control of spinor matterwave. Traditional methods for such suppression include applying a moderate but sufficient quantization field to lift the Zeeman degeneracy [6, 10, 27]. In addition, the \( \Delta m \) spin leakage are naturally suppressed if the Raman transitions are driven by beams with a same circular polarization [54]. These traditional techniques, while showing great success in precision measurements, can be challenging to implement into future compact, flexible devices with SDK control in the \( \omega_{\text{hfs}} \)-limited nanosecond regime.
C. Balanced chirp-alternating SDK scheme

We now demonstrate that the direction of chirps in an adiabatic sequence can be programmed to suppress the coherent accumulation of \(\Delta m\)-leakage. Furthermore, by balancing \(U_{ud}^{(2N)}\) with \(U_{du}^{(2N)}\) as those in Eq. (11), the dynamic phase can be cancelled in \(U_{uddu}^{(2N)}\) to support faithful spinor matterwave phase gate. As to be clarified in the following, both the non-adiabatic spin-leakage suppression and dynamic phase cancellation are supported by time-reversals of the driven spin dynamics, much like those in the traditional spin-echo schemes, but is achieved here in the adiabatic SDK sequence to also ensure the laser intensity-error resilience.

To understand why the accumulation of \(\Delta m\)-leakage error can be partly suppressed by the chirp-alternating \(U_{ud}^{(2N)}(k_R)\) or \(U_{du}^{(2N)}(k_R)\) sequence prescribed in Sec. II E, the \(\Delta m = \pm 2\) leakage itself. In particular, for atom starting in \(|a_m\rangle\) or \(|b_m\rangle\) and subjected to a close-to-ideal adiabatic SDK control, with an \(H_d(r, t)\) Hamiltonian during \(0 < t < \tau_c\) as prescribed in Sec. II E, the \(\Delta m = \pm 2\) transitions driven by \(V'\) are often a result of non-adiabatic couplings among systems with nearly equal energy (for example, the \(m = \pm 1\) subsystems here). By reversing the time-dependence of the Hamiltonian, here with \(H_d(r, t) = H_d(r, \tau a - t)\) within \(\tau a < t < \tau a + \tau_c\), the sign of the non-adiabatic couplings are reversed. Such a sign reversal would lead to complete cancellation of the non-adiabatic transitions if the adiabatic states involved in the couplings are truly degenerate. Here, for atoms being addressed by the cross-linear polarized light, the fact that the \(m\)-spin subsystems are nearly degenerate makes the sign reversal efficient for the suppression of the unwanted leakages.

The simple picture of coherent leakage suppression is verified with numerical simulation in Fig. 5(a,ii) for the chirp-alternating \(U_{ud}^{(2N)}(k_R)\) sequence. Here, in contrast to the \(U_{du}^{(2N)}(k_R)\) case in Fig. 5(a,i), the spin leakage \(\varepsilon_m^{(n)}\) for \(m = 0, 1\) oscillates and is overall efficiently suppressed. Comparing with \(m = 1\) where the leakage is strongly suppressed for even \(n\), the leakage suppression from the \(m = 0\) subsystem follows a more complicated pattern with an approximate periodicity of 4 to 5, suggesting more complex multi-level dynamics (Fig. 1). We further investigate the \(m\)-dependent \(\varepsilon^{(n)}\) dynamics as a function of \(A_R\) in general. The results for \(n = 4N\) are presented in Fig. 5(b-d,ii) to be compared with those in Fig. 5(b-d,i). The \(\Delta m\)-leakage suppression works nearly perfectly for \(m = 1\) subsystem. In fact, we find the chirp-alternation suppresses the accumulation of non-adiabatic transitions between degenerate levels during the multiple adiabatic SDK in general, including those with simple 2-level origin [59, 60].

For \(m = 0, 2\) subsystems, the suppression works fairly well for most \(A_R\) while there are stripes of \(A_R\)-region (around \(A_R = 12\pi\) and \(A_R = 22\pi\) here for example) where the leakage still accumulate with increased \(n = 4N\). The intricate \(A_R\)-dependent \(\varepsilon_m^{(2N)}\) as in Figs. 5(b,h,i)(d,i) suggests that the difference of dynamic phases among \(m = 0, \pm 2\) subsystems by each SDK is large enough to affect the coherent leakage cancellation.

We leave a detailed investigation of the intricate coupling dynamics for future work. Here, to construct faithful spinor matterwave control, we simply combine the \(U_{ud}^{(2N)}\) and \(U_{du}^{(2N)}\) sequence in Eq. (11) to form the balanced chirp-alternating SDK sequence. The idea is to exploit the time-reversal dynamics again to let the dynamic phases by the leakage-suppressing \(U_{du}^{(2N)}(k_R)\) and \(U_{ud}^{(2N)}(k_R)\) cancel each other.

We evaluate the average gate fidelity for realizing the quantum gate for \(U^{(4N)}(k_R)\) (Eq. (8)) with \(\tilde{U}_{uddu}^{(4N)}(k_R)\). The results of \(F^{(4N)}\), with otherwise identical Hamiltonian parameters as those in Fig. 6(a-c,i), are shown in Fig. 6(a-c,ii). Similar to Figs. 6(a-c,i), here the infidelity \(1 - F^{(4N)}\) for the \(\tilde{U}_{uddu}^{(4N)}(k_R)\) sequence closely follow \(\varepsilon^{(4N)}\) for \(U_{ud}^{(4N)}\). Therefore, the improvement is most significant for the \(m = \pm 1\) spin subsystems. For \(m = 0, \pm 2\) subsystems, we also see improved gate fidelity \(F^{(4N)} \geq 95\%\) span a substantial range of intensity for \(4N\) up to 80. Finally, it is interesting to note that for the nearly perfect SDK with \(J_{SDK} \approx 1\), the gate infidelity \(1 - F^{(4N)}\) is dominantly due to the spin leakage \(\varepsilon^{(4N)}\), as demonstrated by the remarkably similar \(1 - F^{(4N)}\) and \(\varepsilon^{(4N)}\) data in Fig. 5 and Fig. 6. This is a result of dynamic phase cancellation in the adiabatic limit, which is supported by both the \(\tilde{U}_{uddu}^{(4N)}\) and \(\tilde{U}_{uddu}^{(4N)}\) sequences, if the \(\pm k_R\) swapping as detailed next is perfect.

D. Robust cancellation of dynamic phase

The numerical results in Fig. 6 demonstrate that precise dynamic phase cancellation can be achieved by pairing \(U_{ud}^{(2N)}(k_R)\) with \(U_{du}^{(2N)}(k_R)\) into the balanced \(\tilde{U}_{uddu}^{(4N)}(k_R)\). In fact, we find that the dynamic phase cancellation in the balanced chirp-alternating scheme is substantially more robust than the standard double-SDK by Eq. (8), as following.

As in Sec. II D, the standard method of dynamic phase cancellation [6] requires perfect \(k_R \leftrightarrow -k_R\) swapping for the successive \(U_K(k_R)\) controls. Practically the k-vector swapping is typically accompanied by a modification of \(E\) intensity ratio. For example, in the retro-reflection setup (Fig. 2(a)), the amplitude of the reflected beam is reduced by a \(\kappa < 1\) factor due to the imperfect reflection, leading to unbalanced dynamic phases \(\psi_D\) associated with \(\tilde{U}(\pm k_R)\) to compromise their cancellation in the standard double-SDK (Eq. (8)). This systematic exists quite generally in retro-reflection setups since the 2-photon shift \(\theta_0\) is sensitive to the laser intensities ratios [25, 54].
In contrast, here we notice that in the $\tilde{U}^{(4N)}_{uudu}(\mathbf{k}_R)$ sequence (Eq. (11)) the dynamic phase by any $\tilde{U}^{(2)}_{ud}(\mathbf{k}_R)$ pair is expected to be cancelled by a $\tilde{U}^{(2)}_{ud}(\mathbf{k}_R)$ pair later. In the adiabatic limit the cancellation is guaranteed, since for free atom starting from any specific 2-level spin state, the sign of the dynamic phase $\delta_{\text{wp}}$ dictates the adiabatic quantum number [53] and thus the sign of the dynamic phase in the adiabatic limit.

To demonstrate the robust dynamic phase cancellation, in Fig. 7 (a,b) we compare $\varphi_D$ according to Eq. (9) for the $\tilde{U}^{(4)}_{ud}(\mathbf{k}_R)$ and $\tilde{U}^{(4)}_{uudu}(\mathbf{k}_R)$ controls, for atom starting from $m = 0, 1$ sub-spaces as examples. The $f_{\text{SDK}}$ values are given in the same plots, with which we see that the high $f_{\text{SDK}}$ is hardly affected by a poor reflectivity $r = |\kappa|^2 = 0.5$. On the other hand, we see that in contrast to Fig. 7(a-b,i) where $r \approx 1$ is required for precise suppression of $\varphi_D$ (blue line), in Fig. 7(a-b,ii) the $\varphi_D$ is largely suppressed so long as $f_{\text{SDK}} \approx 1$, even for a poor $r = 0.5$ as in this experiment. It is worth noting that the residual $\varphi_D$ variation in Fig. 7(a,ii) around $\mathcal{A}_R \approx 12\pi$ is related to coherent spin leakage in the $m = 0, \pm 2$ manifold (Fig. 5(b,d)). For atom starting with $m = 1$ (Fig. 7(b,ii)), the dynamic phase cancellation is essentially perfect even for $r = 0.5$.

**FIG. 7.** Unbalanced dynamic phase $\varphi_D = \varphi_D^{(4)}$ m according to Eq. (9) vs pulse area $\mathcal{A}_R$ for $m = 0$ (a) and $m = 1$ (b) subjected to $\tilde{U}^{(4)}_{ud}(\mathbf{k}_R)$ (i) and $\tilde{U}^{(4)}_{uudu}(\mathbf{k}_R)$ (ii) controls. The corresponding 2-photon detuning profiles are illustrated on the top. The data are simulated for corresponding 2-photon detuning profiles are illustrated on the top. In all the graphs the red dashed lines give $f_{\text{SDK}}$ for single kicks where different $r$-curves closely overlap. The 2-photon detuning profiles are illustrated on the top.

### E. Area-enhancing atom interferometry

So far in this section, we have shown that matterwave phase gate prescribed by the ideal double-SDK (Eq. (8)) can be faithfully implemented by the balanced chirp-alternating sequence $\tilde{U}^{(4N)}_{uudu}(\mathbf{k}_R)$, to coherently shift any $|a_m\rangle$, $|b_m\rangle$ components of hyperfine spinor matterwave with opposite $\pm 4N\hbar\mathbf{k}_R$ momentum within nanoseconds. For control parameters in this experimental demonstration, our numerical results already suggest high gate fidelity with efficient suppression of the coherent spin leakage and inhomogeneous dynamic phase. In future work, by increasing $\Delta_\sigma/T$ and $\Delta_e/\Delta_{hfs,e}$ ratios and the laser intensities $I_{1,2}$ in proportion, the residual imperfections can be further suppressed to meet the exquisite requirements in the applications of quantum information processing [11, 12, 16, 17] and quantum enhanced atom interferometry [18–21].

Here, to demonstrate the utility of the adiabatic SDK sequence for precision measurements, we numerically investigate a simple atom interferometry scheme [3, 6, 10] where an "enclosed area" $A$ is enhanced by the $\tilde{U}^{(4N)}_{uudu}(\pm \mathbf{k}_R)$ sequences. As in the Mach-Zehnder configuration in Fig. 8(a), we consider the two spinor matterwave components forming a loop to interfere at $t = 2T$ (the dashed lines): when the duration of the pulsed rotations are negligibly short compared to the "interrogation time" $T$, then the spatial-temporal "area" enclosed by the loop is easily evaluated as $A = v_R T^2$ with $v_R = \hbar k_{\text{eff}}/M$. Importantly, the "enclosed area" $A$ is an integrated phase-space separation between the two interfering paths of matterwave, which is often proportional to the differential action experienced by the atom along the two paths, such as by a gravitational force or a Coriolis force to be read out interferometrically. To achieve such large "area" $A$ as possible within a measurement time $T$ is thus of essential importance to precision measurements with light pulse atom interferometry [3, 34, 61].

More specifically, the enclosed area is defined as $A = \int_0^{2T} \Delta z(t)dt$ during a 3-pulse Raman interferometry sequence by integrating the relative displacement $\Delta z(t)$ between the two matterwave diffraction paths under the three operations as splitter ($t = 0$), mirror ($t = T$) and combiner ($t = 2T$). We generally refer the idealized local spin rotations as $R_\omega(\theta) = \cos(\theta/2) 1 + i \sin(\theta/2) (e^{i\sigma_+} e^{-i\sigma_-})$ for the Raman interferometer, for any spin state within $\{|a_m\rangle, |b_m\rangle\}$. Here $\varphi = \mathbf{k}_R \cdot \mathbf{r}$ is the local Raman optical phase. Notice the spatial-dependent $R_\omega(\theta)$ rotation can in principle be generated by the Eq. (4) Hamiltonian [62] as phase-coherent "half" and "full" kicks. The splitter and mirror operations in the standard light Raman interferometer can then be conveniently expressed as $R_1 = R_\omega(\pi/2)$, $R_2 = R_\omega(\pi)$ and $R_3 = R_\omega(\pi/2)$ respectively to manipulate the spin states while imparting the $\pm \hbar k_R$ photon recoil momentum.

We now consider enhancing the enclosed area $A$ of the standard 3-pulse Raman interferometer with the chirp-
FIG. 8. Enhancing the enclosed area of an atom interferometer with four $\tilde{U}_{uddu}(\pm k_R)$ SDK sequences. (a): Schematic of the interferometry scheme. The orange dashed lines at $t = 0, T, 2T$ represent regular Raman interferometry controls for $R_1 = R_p(\pi/2)$ splitter, $R_2 = R_p(\pi)$ mirror and $R_3 = R_p(\pi/2)$ combiner of the spinor matterwave respectively with $\varphi = k_R \cdot r$. The atom in $|b\rangle$ and $|a\rangle$ states is represented by the blue and red lines. The four thick purple-colored vertical lines at $t = \tau_1, T - \tau_1, T + \tau_1, 2T - \tau_1$ with red curved arrows represent $\tilde{U}_1 = \tilde{U}_{uddu}(k_R)$, $\tilde{U}_2 = \tilde{U}_{uddu}(-k_R)$, $\tilde{U}_3 = \tilde{U}_{uddu}(2k_R)$ and $\tilde{U}_4 = \tilde{U}_{uddu}(-k_R)$ sequences respectively. We consider $\tau_1 < T$. By properly choosing $\tau_1/T$ ratio, spurious interference by multiple imperfect controls can be suppressed, and are not included in the simulations. (b-d): The interferometry phase offset $\delta \Phi$ and contrast $C$ as a function of SDK number $n = 4N$ and pulse area $\Delta R$. Here the single-photon detuning is chosen as $\Delta_\nu = 3.3 \, \omega_{hfs,c}$. The simulations average over unpolaredized $m = -2, -1, 0, 1, 2$ states, and include $\Gamma_\nu = 0.017 \, \omega_{hfs,c}$ as for the case of $^{85}\text{Rb}$. The phase offsets and interferometry contrasts locally averaged over a 50% intensity are given in (c,d). The data in Fig. (e-g) are similar to Fig. (b-d), but with an increased single-photon detuning of $\Delta_\nu = 6.6 \, \omega_{hfs,c}$.

alternating SDK sequence. In particular, we consider the Fig. 8(a) scheme with the spinor matterwave diffraction paths marked with solid lines: a $\tilde{U}_1 = \tilde{U}_{uddu}(k_R)$ is first applied at $t = \tau_1$ to increase the momentum displacement between the two interfering paths from $\Delta \mathbf{p} = h \mathbf{k}_R$ by $R_p(\pi/2)$ to $\Delta \mathbf{p} = (2 \times 4N + 1)h \mathbf{k}_R$ with the spin-dependent kicks. This $\Delta \mathbf{p}$-enhancement is followed by an opposite $\tilde{U}_2 = \tilde{U}_{uddu}(-k_R)$ at $t = T - \tau_1$ before the $R_3$-operation to recover the initial $\Delta \mathbf{p}$. To ensure that the interfering paths spatially overlap at $t = 2T$, an additional pair of opposite momentum boosts, $\tilde{U}_{3,4}$ are applied at $t = T + \tau_1$ after the $R_2$ and $t = 2T - \tau_1$ before the $R_3$ operation respectively. By properly choosing the $\tau_1/T$ ratio, spurious interference by imperfect $R_{1,2,3}$ and $\tilde{U}_{1,2,3,4}$ controls can be suppressed [63, 64]. With $T \gg \tau_1$, the enclosed area of the resulting interfering loop is enhanced to $A' = (2 \times 4N + 1)A$.

For the numerical simulation, we consider at $t = 0$ the atomic state to be initialized at certain $|b_m\rangle$ and subjected to the $\tilde{U}_{uddu}(k_R)$-enhanced 3-pulse interferometry sequence. The output atomic state, right before the final matterwave combiner $R_3$, can then be written as $|\psi_m(r, 2T^-)\rangle = U_{AI} |c_m\rangle$, with $U_{AI} = U_f(\tau_1)U_{UU}(T - 2\tau_1)U_{UU}(T - 2\tau_1)U_{UU}(T - 2\tau_1)U_{UU}(T - 2\tau_1)$ to be $r$-dependent. Here $U_f(t)$ designates free propagation of matterwave for time $t$. The $R'_1 = U_f(\tau_1)R_1, R'_2 = U_f(\tau_1)R_2U_f(\tau_1)$ take into account the free propagation of matterwave between the standard $R_j$ and area-enhancing $U_j$ sequences. We numerically evaluate $|\psi_m(z, 2T^-)\rangle$ for 1D spinor matterwave between $0 < z < \lambda/2$, as described in Appendix B. To focus on the performance of SDK, we set $R_1,3$ as perfect $\pi/2$ pulses and $R_2$ as perfect $\pi$ mirror pulse respectively. A further simplification sets $k_R = 0$ for the idealized $R_{1,2,3}$ controls, with which we numerically evaluate $|\Sigma_j\rangle = \langle \psi_m(z; 2T^-)|\Sigma_j|\psi_m(z; 2T^-)\rangle_{z,m}$ for an initially unpolared atomic sample right before the $R_3$ operation. Here $\Sigma_j = \sum_{m' = \frac{1}{2} - \frac{3}{2}} |\sigma_j^{(m')}\rangle$ are summed over all $m$ sub-spins for Pauli matrices with $j = x,y,z$. $\Sigma_j$ corresponds to observables of experimental measurements in which Zeeman sublevels are not resolved, as in most atom interferometry experiments with hyperfine state-dependent fluorescence readouts [34].

With $U_f$ chosen as free 1D propagation, the values of $\tau_1$ and $T$ only affects contributions of spurious interfering paths into the final readouts [63, 64] in the simulation. With $R_{1,2,3}$ set as ideal, the spurious interfering paths are from imperfect $\tilde{U}_{1,2,3,4}$ diffractions only. Notably, since successive adiabatic SDKs here within each $U_f$ last merely tens of nanoseconds, the spatial displacements among the spurious interfering paths are negligibly small comparing with the typical coherence length of cold atom samples, and therefore do not alter the matterwave dynamics [65, 66]. We have randomly sampled the atomic initial position and velocity to numerically verify that the residual spurious interference are indeed suppressible. Practically, to generate the results in Fig. 8(b-d) with all spurious interference removed in an efficient manner, we simply apply a digital filter to remove unwanted diffraction orders after each $U_f$ sequence.

We are particularly interested in the interferometry
contrast $C$ and diffraction phase offset $\delta \Phi$. The contrast $C$ decides the quality of the final matterwave interference fringes. The phase offset $\delta \Phi$, stemming from the unbalanced dynamic phase by the four $U_{uddu}$ sequences, enters the interferometry readout as systematic bias against any precision measurements or controls. Numerically, we conveniently evaluate the interferometry contrast as $C = \sqrt{(\Sigma_x)^2 + (\Sigma_y)^2}$. The diffraction phase offset is instead evaluated as $\delta \Phi = \text{arg} (|\Sigma_x| + i|\Sigma_y|) - \Phi_0$ with $\Phi_0$ to be the relative phase between $|a_m\rangle$ and $|b_m\rangle$ right after the ideal $R_1$ splitter. Typical numerical results are presented in Figs. 8(b-d). The simulation is again performed on the $^{85}\text{Rb}$ D1 line, here with spontaneous emission included. For Figs. 8(b-d) on the left panel, the control laser parameters are chosen close to this experimental work, with $\Delta_\epsilon = 3.3$ $\omega_{\text{hfs,g}}$ so both $\varepsilon_{\text{sp}}$ and $\varepsilon_{\Delta m}$ are quite substantial. Nevertheless, we find $C > 0.5$ with $\delta \Phi < 0.01$ after four $n = 4N = 12$ chirp-alternating SDKs are applied for a 25-fold enhancement of interferometry enclosed area. Here, to avoid excessive spontaneous emission and coherent leakage (Fig. 5(b-d)), The peak $A_R = 6 - 8\pi$ needs to be chosen (Fig. 8(b-d)). On the other hand, by doubling the single photon detuning to $\Delta_\epsilon = 6.6$ $\omega_{\text{hfs,g}}$ (with laser intensity increased in proportion to maintain the Raman Rabi frequency), $\varepsilon_{\text{sp}}$ are halved, while the impact of $\varepsilon_{\Delta m}$ leakage are dramatically suppressed in the $U_{uddu}$ sequence (Figs. 8(e-g)). The further detuned $U_{uddu}$ sequence should thus support up to 50-fold enhancement of interferometry enclosed area, with spontaneous-emission-limited $C > 0.5$ contrast and negligible $\delta \Phi$ offset.

V. DISCUSSIONS

Significant aspects of advanced quantum technology today are based on controlling alkaline atoms through their center-of-mass motion and ground-state hyperfine interaction. The two long-lived degrees of freedom can be entangled optically by transferring photon recoil momentum with Raman excitations. The tiny atomic recoil effect can be amplified by repetitive application of such excitations. The spin-dependent large momentum transfer is expected to improve the scalability for precision measurements in atom interferometry [3, 6, 10] and for quantum information processing with trapped ions [5, 11–13, 17]. Practically, unlike interrogating microscopically confined single ions where Raman excitation with multiple-THz single-photon detuning is feasible [8, 39], addressing larger samples prefers efficient excitations at moderate single-photon detunings. The seemingly unavoidable imperfections associated with spin-leakages and dynamic phases need to be managed in non-traditional ways, for achieving the high speed, high fidelity control with intensity-error resilience required by the next generation quantum technology [16–21].

In this work, we have demonstrated a novel configuration of adiabatic SDK implemented on an optical delay line, which is able to reach the speed limit of Raman SDK control [5, 39], featuring robust intensity-error resilience, while maintaining various advantages of optical retro-reflection established for precision atom interferometry. The experimental characterization of the technique is limited to incoherent observables, but we clarify in Sec. IV that high precision phase gates enabling spin-dependent large momentum transfer can be efficiently realized at the moderate single-photon detuning, by properly programming the chirp direction to suppress the accumulation of coherent errors. We have provided numerical evidence that the chirp-balanced SDK scheme support faithful, parallel $\Delta m = 0$ control of multi-Zeeman spinor matterwave, with giant spin-dependent forces applied within nanoseconds to rapidly shift the phase-space spin separation, even with the $\sim$10 mW laser power as in this work. Since within nanoseconds various low-frequency noises including those due to matterwave dispersion are negligible for cold atoms, we expect accurate implementation of the full scheme to be benchmarked in future interferometric measurements.

The performance of the adiabatic SDK demonstrated in this experimental work can be substantially improved by equipping programmable optical waveform generation with a Watt-level peak power [36]. In that case, a ten-fold increase of $\Delta_\epsilon$ to 100 GHz can substantially suppress $\varepsilon_{\text{sp}}$ and $\varepsilon_{\Delta m}$ errors to 0.1% level, while a ten-fold reduction of SDK pulse duration can be achieved at the same time. For SDK within a few nanoseconds, a meter-long delay line is able to spatially resolve counter-propagating pulses to flexibly drive bidirectional SDKs. With the chirp-balanced scheme, the compact device should flexibly support spin-dependent coherent control of hyperfine matterwave with unprecedented speed and precision. Finally, we remark that for the spinor matterwave control with the delay-line based SDK scheme, as in Fig. 2, the extra dynamic phases by the pre- and post-pulses need to be precisely compensated. Similar to the frequency domain Stark shift compensation [61], for the nanosecond SDKs here one may fire additional pulses with suitable single-photon detunings to trim the overall dynamic phase. Within nanoseconds, cold atoms hardly move to change the local laser intensity. We therefore expect the dynamic phase compensation to function well in the time domain.

ACKNOWLEDGEMENTS

We are grateful to Prof. Yiqiu Ma for insightful comments to the manuscript, and to Prof. Xiaopeng Li and Prof. Haidong Yuan for helpful discussions. We acknowledge support from National Key Research Program of China under Grant No. 2016YFA0302000 and No. 2017YFA0304204, from NSFC under Grant No. 12074083.
Appendix A: Full Hamiltonian

The theoretical analysis as well as numerical simulation in this work is based on the full light-atom interaction Hamiltonian on the D1 line. Following the notation in the main text, the effective, non-Hermitian Hamiltonian is written as

\[ H_{\text{eff}}(\mathbf{r}, t) = \hbar \sum_{c} \left( \omega_c - \omega_{e\delta} - i \Gamma_c / 2 \right) \sigma^{c_i e_i} + \]

\[ \hbar \sum_{c=a,b} (\omega_c - \omega_{g\delta}) \sigma^{e_m c_m} + \]

\[ \frac{\hbar}{2} \sum_{c=a,b} \sum_{i} \Omega_i^c \sigma^{e_m c_m}(\mathbf{r}, t) + \text{h.c.} \]  

(A1)

Here \( \omega_{e\delta}, \omega_{g\delta} \) are decided by the energy of reference level in the excited and ground state manifolds respectively, chosen as the top hyperfine levels in this work. The laser Rabi frequency, \( \Omega_i^c \) is accordingly written in the \( \omega_{e\delta}, \omega_{g\delta} \) frame under the rotating wave approximation. \( \mathbf{d} \) to be the atomic electric dipole operator. The \( \sigma^{e_m c_m} = |a_m\rangle \langle e_i| \) \( \sigma^{e_i e_k} = |e_k\rangle \langle e_i| \) are the raising and lowering operators between states \( |a_m\rangle \) and \( |e_i\rangle \). Similar \( \sigma \) operators are defined for all the other \( |a_m\rangle, |b_n\rangle \) and \( |e_i\rangle \) state combinations.

The full \( H_{\text{eff}} \) in Eq. (A1) is rewritten as Eq. (4) in the main text. For weak off-resonant pulses and to facilitate understanding of ground-state Raman interaction, we can also adiabatically eliminate the excited states to approximately have

\[ H_{\text{eff}}(\mathbf{r}, t) \approx \]

\[ \hbar \sum_{c} \sum_{j=1,2} \frac{\Omega_{a_m e_i}^{j} \sigma^{j a_m a_n} + \Omega_{b_m e_i}^{j} \sigma^{j b_m b_n}}{4(\nu_j - \omega_{e\delta})} + \]

\[ \hbar \sum_{c=a,b} \sum_{j} \frac{\Omega_{a_m e_i}^{j} \sigma^{j a_m a_n} + \Omega_{b_m e_i}^{j} \sigma^{j b_m b_n}}{4(\nu_j - \omega_{e\delta})} + \sum_{c=a,b} \sum_{j} \frac{\Omega_{a_m e_i}^{j} \sigma^{j a_m a_n} + \Omega_{b_m e_i}^{j} \sigma^{j b_m b_n}}{4(\nu_j - \omega_{e\delta})} \]  

with the convention of summing over repeated indices \( m, n, l \). The single photon detuning is defined as \( \Delta_c = \nu_i - \omega_{e\delta} = \nu_j - \omega_{e\delta} \).

Appendix B: Numerical model

We numerically simulate the evolution of 1D spinor matterwaves with full D1 light-atom interactions \([\text{67, 68]}\) driven by the counter-propagating Raman pulses as in Fig. 1 and 2. To account for radiation damping, we follow a stochastic wavefunction method \([\text{69]}\) to evaluate the wavefunction \( |\psi(\mathbf{r}, t)\rangle \) for atom at location \( \mathbf{r} \) under the non-Hermitian Hamiltonian \( H_{\text{eff}} \) (see Eq. (A1)) with non-Hermitian part \( \hbar \sum_c i \Gamma_c \sigma^{e_i e_i} / 2 \) in the first line. Here \( \Gamma_c \) is the natural linewidth of the D1 line. The simulations treat both the internal and external motion of the spinor matterwave quantum mechanically. For the purpose, we sample \( |\psi(\mathbf{r}, t)\rangle \) densely over a uniform grid within \( 0 < z < \lambda / 2 \) and sparsely in the \( x - y \) plane, and follow a split-operator method to evaluate internal/external atomic motion numerically with interleaved steps. Taking advantage of the short \( \tau_c \) for single SDK, the internal state dynamics is evaluated within a single step with frozen external motion under a local \( |\psi(\mathbf{r})\rangle \) basis, with atomic position \( \mathbf{r} \) treated as a parameter of \( H_{\text{eff}} \). The evaluation of observables later is normalized by \( \mathcal{N} = \sum_{r} \langle \psi(\mathbf{r}, t = 0)|\psi(\mathbf{r}, t = 0)\rangle \), with corrections from stochastic contributions to be discussed shortly. Between SDKs, a Fourier transform along \( \mathbf{e} \) can be performed to evolve the free-flying spinor matterwave along \( z \) if necessary. To save computation resources, the relatively simple atomic dynamics in the \( x - y \) plane is ignored. To evaluate momentum distribution of spinor matterwave, we simply perform a Fourier transform to the space dependent \( \langle c_m|\psi(\mathbf{r}, t)\rangle \) for any specific spin state \( |c_m\rangle \).

Beyond the coherent evolution, the simulation complexity is substantially reduced by skipping the evaluation of stochastic trajectories heralded by a single “quantum jump” \([\text{47, 48]}\). Specifically, after each pulsed interaction, the trajectories suffering a quantum jump are simply assumed to repopulate \( \{|a\}, |b\} \) in a uniform manner with properly shifted photon recoil momentum. Without further evolution, these trajectories contribute to the evaluation of incoherent, single-time observables such as hyperfine population and photon momentum transfer. The overall probability of spontaneous emission is determined by the norm of the final wavefunction, \( \varepsilon_{sp} = 1 - 1 / N \sum_{r} \langle \psi(\mathbf{r}, \tau_{tot})|\psi(\mathbf{r}, \tau_{tot})\rangle \), after a total evolution time \( \tau_{tot} \). The simplification is generally justified for evaluating coherent observables of interest, since the expectation values shifted stochastically lead to zero coherent contributions. For the incoherent observables such as average photon momentum and hyperfine population, the simplification is supported by the simple D1 structure under consideration here \([\text{67]}\), where a single spontaneous emission effectively randomizes the following Raman interaction dynamics.
Appendix C: A Markovian model for $f_{SDK}$ estimation

For atoms subjected to multiple SDKs, the dynamics of spinor matterwave that deviates from the ideal control can be depicted as diffusing in a “momentum-lattice” [15]. Our numerical simulation suggests that with fair efficiency of single adiabatic pulse to achieve hyperfine transfer efficiency of $f_R > 95\%$, the resulting average momentum $p_n$ and population $\rho_{aa/bb,n}$ roughly follow a simple Markovian model. The model assumes that both the momentum and population transfer from the next kick are decided by the present population difference $\rho_{aa} - \rho_{bb}$ only. The details of the Markovian model is described as follows.

Suppose that after $n$ kicks, the atomic ensemble is with momentum $p_n$ (in unit of $\hbar k_R$) and population contrast $C_n \equiv |\rho_{aa,n} - \rho_{bb,n}|$, then the next kick will impart momentum as

$$\Delta p_{n+1} = p_{n+1} - p_n = f_0 (1 - \varepsilon_{sp}/2) C_n,$$  \hspace{1cm} (C1)

where $f_0$ is the hyperfine population transfer efficiency in absence of the spontaneous emission. Here we have assumed that during the single pulse process, the spontaneous emission occurs with a uniform distribution of probability, thus the associated population recycled to the ground states acquires half of $\hbar k_R$ momentum on average.

Similarly, the population distribution can be written as

$$\rho_{aa,n+1} = (1 - \varepsilon_{sp}) \left[ (1 - f_0) \frac{1 + C_n}{2} + f_0 \frac{1 - C_n}{2} \right] + \varepsilon_{sp}/2,$$

$$\rho_{bb,n+1} = (1 - \varepsilon_{sp}) \left[ f_0 \frac{1 + C_n}{2} + (1 - f_0) \frac{1 - C_n}{2} \right] + \varepsilon_{sp}/2,$$  \hspace{1cm} (C2)

so that

$$C_{n+1} = (1 - \varepsilon_{sp}) (2f_0 - 1) C_n.$$  \hspace{1cm} (C3)

We define Raman transfer efficiency as $f_R = f_0 (1 - \varepsilon_{sp}/2)$. When both $f_0$ and $1 - \varepsilon_{sp}$ are close to unity, Eqs. (C1)-(C3) can be approximated as

$$p_{n+1} - p_n = f_R C_n,$$

$$C_{n+1} = (2f_R - 1) C_n.$$  \hspace{1cm} (C4)

With the recursion relations by Eq. (C4), we arrive at

$$p_n = f_R - \frac{1 - (2f_R - 1)^n}{1 - (2f_R - 1)},$$

$$C_n = (2f_R - 1)^n.$$  \hspace{1cm} (C5)

Finally, we remark that for the Raman SDK, there is a slight difference of spontaneous emission loss for single kicks between the $a \to b$ and $b \to a$ process. As we consider repetitive SDK with $n$ up to a quite large number (e.g. $n_{\text{max}} = 25$ in our experiment), we effectively set a same $\varepsilon_{sp}$ parameter for the opposite population transfer processes.

Appendix D: Absorption imaging analysis

In Sec. III we have introduced the double imaging technique. This section provides details on deriving recoil momentum $p_n$ and population transfer $\rho_{aa}$ from the imaging data.

Our atomic sample is prepared in a cross-dipole trap with slight asymmetry. When deriving the momentum transfer from the double images as in Fig. 3, we found that neither before nor after the time-of-flight, the absorption profile can fit perfectly to a 2D Gaussian. In addition, due to the relatively short exposure time of 20 $\mu$s to the weak probe ($s = 1$), there are substantial photon shot noise in single-shot images. To faithfully retrieve central position and atom number from each pair of double-image, we take the following procedure. First, we repeat a same type of measurements for $N = 80$ shots, and do a principle components analysis to all pairs of double-images after background subtractions. The first three components are kept for the following analysis. We then do a 2D Gaussian fit, expanding the fitted Gaussian profiles to 1.5 times the waist into a wide enough stepwise mask. The population ratios $\rho_{aa/bb}$ are evaluated by the ratio of total counts between the two images within the mask, where the center-of-mass (COM) positions $z_1$ and $z_2$ are also directly evaluated.

We note that the first image is contributed by atoms at $F = 3$ (“$a_{m}$”) states only. In other words, the atoms kicked to the “visible” (or “invisible”, depends on odevity of kicking number $n$) hyperfine levels are post-selected. Since there is a small interval $\tau_{p,1}$ between SDK and the first image, there is a bias to the COM position $z_1$ of the whole cloud due to this post-selection. To correct for the bias, we rewrite the position difference as $z_2 - z_1 = \tilde{v}_n \tau_{t o f} + \rho_{bb, n} \tilde{v}_b, n \tau_{p,1}$. Here $\tilde{v}_n$ and $\tilde{v}_b, n$ are the mean velocity for the whole atomic ensemble and for the atoms in $F = 2$ after $n$ SDKs, respectively. The correction to the post-selection induced velocity bias is then given by

$$\tilde{v}_n = v_n \left( 1 - \rho_{bb, n} v_{bb, n} \frac{\tau_{p,1}}{\tau_{t o f}} \right) = v_n \left( 1 - \xi (n, v_n) \frac{\tau_{p,1}}{\tau_{t o f}} \right).$$  \hspace{1cm} (D1)

Here, $\xi (n, v_n)$ depends on the relation between $v_n$ and $\rho_{bb, n}, v_{bb, n}$. The relation can be approximated with the model in Appendix C. With $\tau_{p,1} \sim 15$ $\mu$s in our experiment, this correction is typically $1 - \tilde{v}_n/v_n \sim \pm 5\%$ (The $\pm$ signs depend on $n$), which impacts $f_R$ at 3% level. The correction is model-dependent. We correct for the bias in our final estimation of $f_{SDK}$ and leave 3% as the dominant uncertainty in our $f_{SDK}$ estimation.

Appendix E: Impact of mirror optical loss to $f_{SDK}$

Numerical simulations suggest that the reflectivity $r = |\kappa|^2$ of the retro-reflection mirror (Fig. 2) affect $f_{SDK}$ slightly. To investigate the effect, we sample $0 < r < 1$ during the simulation, and calculate Raman transfer effi-
FIG. A1. (a) Numerical simulations for $f_R$ and $f_{SDK}$ vs Raman pulse area $A_R$ for different reflectivity $r = |k|^2$ of the retro-reflection mirrors. The red bars show estimated distribution of atoms subject to different pulse areas under the Gaussian beam illumination in this experiment. (b) $n(A_R)$-weighted average $f_R$ and $f_{SDK}$ vs reflectivity $r$. The experimentally measured $f_R$ and inferred $f_{SDK}$ are marked with error bars.

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