Composite pulses for interferometry in a thermal cold atom cloud

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Atom interferometric sensors and quantum information processors must maintain coherence while the evolving quantum wavefunction is split, transformed and recombined, but suffer from experimental inhomogeneities and uncertainties in the speeds and paths of these operations. Several error-correction techniques have been proposed to isolate the variable of interest. Here we apply composite pulse methods to velocity-sensitive Raman state manipulation in a freely-expanding thermal atom cloud. We compare several established pulse sequences, and follow the state evolution within them. The agreement between measurements and simple predictions shows the underlying coherence of the atom ensemble, and the inversion infidelity in a ~ 80 µK atom cloud is halved. Composite pulse techniques, especially if tailored for atom interferometric applications, should allow greater interferometer areas, larger atomic samples and longer interaction times, and hence improve the sensitivity of quantum technologies from inertial sensing and clocks to quantum information processors and tests of fundamental physics.

Emerging quantum technologies such as atom interferometric sensors[1], fountain atomic clocks[2] and quantum information processors[3] rely upon the precise manipulation of quantum state superpositions, and require coherence to be maintained with high fidelity throughout extended sequences of operations that split, transform and recombine the wavefunction. In practice, however, inhomogeneities lead to uncertainty in the rates and phase space trajectories of these operations. To reduce the sensitivity of the intended operation to variations in laser intensity, atomic velocity, or even gravitational acceleration[4], several approaches have been proposed, from quantum error correction[5] to shaped pulses[6] and rapid adiabatic passage[7–9]. Just as squeezing does for an individual wavefunction[10], these techniques aim to reduce the uncertainty projected within an ensemble distribution upon the parameter of interest.

NMR spectroscopists have over many years developed ‘composite pulse’ techniques to compensate for systematic variations in the speed and trajectory of coherent operations, and thus refocus a quantum superposition into the desired state[11,12]. The various pulse sequences differ in their tolerance of ‘pulse length’ (or coupling strength) and ‘off-resonance’ errors and correlations between them, and in the operations for which they are suitable and the properties whose fidelity they protect. All are in principle applicable to the coherent control of any other two-state superposition, and such techniques have been applied to the manipulation of superconducting qubits[14], diamond NV colour-centres[15], trapped ions[16–20], microwave control of neutral atoms[21–25], and even the polarization of light[26].

Perhaps the simplest composite pulse sequence, based upon Hahn’s spin-echo[27], inserts a phase-space rotation between two halves of an inverting ‘π-pulse’ to compensate for systematic variations in the coupling strength or inter-pulse precession rate. A number of researchers have applied such schemes to optical pulses in atom interferometry, using the π-pulse also to ensure proper path overlap analogous to the mirrors of a Mach-Zehnder interferometer. Using stimulated Raman transitions from a single Zeeman substate in a velocity-selected sample of cold Cs atoms, Butts et al.[28] extended this scheme by replacing the second π/2-pulse with one three times as long, thus forming a WALTZ composite pulse sequence[29] that, with appropriate optical phases, is tolerant of detuning errors and hence the Doppler broadening of a thermal sample. Following the proposal of McGuirk et al.[30] that composite pulses could withstand the Doppler and field inhomogeneities in ‘large-area’ atom interferometers, in which additional π-pulses increase the enclosed phase space area to raise the interferometer sensitivity, Butts et al. showed that the WALTZ pulse increased the fidelity of such augmentation pulses by around 50%.

In this paper, we use velocity-sensitive stimulated Raman transitions to compare the effectiveness of several established pulse sequences upon an unconfined sample of 85Rb atoms, distributed across a range of Zeeman substates, after release from a magneto-optical trap. We explore the CORPSE, BB1, KNILL and WALTZ sequences, determine both the detuning dependence and the temporal evolution in each case, and show that the inversion infidelity in a ~ 80 µK sample may be halved from that with a basic ‘square’ π-pulse. Comparison with simple theoretical predictions shows the underlying coherence of the atomic sample, and suggests that if cooled towards the recoil limit such atoms could achieve inversion fidelities above 99%. Our results demonstrate the feasibility of composite pulses for improving pulse fidelity in large-area atom interferometers and encourage the development of improved pulse sequences that are tailored to these atomic systems[31]: they also open the way to interferometry-based optical cooling schemes such as those proposed in[32] and[33].
RESULTS

We explore a popular atom interferometer scheme, used to measure gravitational acceleration [1, 24], rotation [25] and the fine-structure constant [26], in which stimulated Raman transitions [27] between ground hyperfine states provide the coherent ‘beamsplitters’ and ‘mirrors’ to split, invert and recombine the atomic wavepackets; motion, acceleration or external fields then induce phase shifts between the interferometer paths that are imprinted on the interference pattern at the interferometer output. Our experiments are performed on a cloud of about $2 \times 10^7$ $^{85}$Rb atoms with a temperature of 50–100 µK, and the 780 nm Raman transition is driven between the $F = 2$ and $F = 3$ ground states (Figure 1(a)). The two laser fields are detuned (Δ) from single-photon hyperfine splitting, ∆ = 10 GHz, and the counterpropagating Raman beams are opposite-circularly polarised $\sigma^+$ and $\sigma^-$. The relative transition strengths, calculated from the Clebsch-Gordan coefficients, normalised to the 0–0 transition, are given for each route.

via the Raman routes shown in Figure 1(b), where, for angular momentum to be conserved, $\Delta m_F = 0$ for the Raman transition regardless of the quantisation axis, but the different coupling strengths lead to different light shifts for different $m_F$ sub-states. With orthogonal linear polarizations ($\pi^+ - \pi^-$), which correspond to superpositions of $\sigma^+$ and $\sigma^-$ components, the two $\Delta m_F = 0$ components add constructively and the $m_F$ dependence of the light shift disappears. For parallel linear polarizations (e.g. $\pi^+ - \pi^+$), the $\Delta m_F = 0$ components cancel.

The Raman coupling strengths and resonance frequencies depend upon the hyperfine sub-state (shown in Figure 1(b)), the atom’s velocity and, via the light shift, the intensity at the atom’s position within the laser beam. These inhomogeneities lead to systematic errors in the manipulation processes and hence dephasing of the interfering components, limiting the interferometric sensitivity. A common solution [28] is to spin-polarise the atomic ensemble into a single Zeeman sub-state, and pre-select a thermally narrow ($T < 1 \mu$K) portion of its velocity distribution before the Raman pulses are applied. Both of these processes however reduce the atom number and hence also the signal-to-noise of the interferometric measurement. Adiabatic rapid passage offers inhomogeneity-tolerant population transfer from a defined initial state, but is inefficient for the recombination of superpositions of arbitrary phase [29, 40]. Composite pulses can in contrast operate effectively, in the presence of inhomogeneities, upon a variety of superposition states.

The effects of experimental inhomogeneities are apparent in Figure 3, which shows Rabi flopping in a Zeeman-degenerate atom cloud at $\Omega_{\text{eff}} \approx 2 \pi \times 200$ kHz, where the mean upper hyperfine state population $|v_2|^2$ is measured as a function of Raman pulse length $t$. The atoms dephase almost completely within a single Rabi cycle, and the upper state population settles at a transfer fraction of 0.28. The peak transfer fraction is about 0.5.
features but contribute little to the overall dephasing.

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sidity inhomogeneities are included at the observed level of as in Figure 2, with parameters given in Table II. INTen-
tion corresponding to a superposition of two Gaussians state, with uniform illumination and a velocity distribu-
equally-populated m states are given in the Methods section. The simulated effect of our laser pulses upon the atom cloud: details are given in the Methods section. The Bloch vector trajectory, starting from |ψ⟩ as a function of the interaction time; phase and de-
sired rotation, azimuth and polar angles θ, φ and α are achieved by setting the interaction time, phase and de-
tuning of the control field respectively. Resonant control fields cause rotations about axes through the Bloch sphere equator (α = 0), and result in Rabi oscillations in the state populations as functions of the interaction time; off-resonant fields correspond to inclined axes.

The pulse sequences explored here all use fields that are set to be resonant for stationary atoms, so we assume α to be zero and write θ0 ≡ U(θ, φ, 0) to represent a rotation defined by the angles θ and φ (written in degrees). A sequence of such rotations is written as θ(1)φ(1)θ(2)φ(2)... where chronological order is from left to right. Two pulses commonly used in atom interferometry are the ‘mirror’ π pulse, represented as the rotation 180φ, and the ‘beamsplitter’ π rotation defined by the angles θ and φ (written in degrees). A sequence of such rotations is written as θ(1)φ(1)θ(2)φ(2)... where chronological order is from left to right. Two pulses commonly used in atom interferometry are the ‘mirror’ π pulse, represented as the rotation 180φ, and the ‘beamsplitter’ π pulse, represented as 90φ.

For each set of experimental results, we also show the simulated effect of our laser pulses upon the atom cloud: details are given in the Methods section. The solid curves of Figure 3 are numerical simulations for equally-populated mF sub-states of the F = 2 hyperfine state, with uniform illumination and a velocity distribution corresponding to a superposition of two Gaussians as in Figure 2 with parameters given in Table II. Intensity inhomogeneities are included at the observed level of ∼7% and, with 1/f noise (green curve), wash out minor features but contribute little to the overall dephasing.

Bloch sphere notation

Coherent operations in atom interferometry may be visualised upon the Bloch sphere, whereby the pure quantum states |1⟩ and |2⟩ lie at the poles and all other points on the sphere describe superpositions with various ratios and phases [111]. Raman control field pulses correspond to trajectories of the two-level quantum state vector |ψ⟩ on the surface of the sphere. For constant intensities and frequencies, these are unitary rotations, and the unitary rotation propagator acting on |ψ⟩ takes the form [12]

$$U(\theta, \phi, \alpha) = \cos \left(\frac{\theta}{2}\right) I - i \sin \left(\frac{\theta}{2}\right) \left[\sigma_x \cos(\phi) \cos(\alpha) + \sigma_y \sin(\phi) \cos(\alpha) + \sigma_z \sin(\alpha)\right],$$

(1)

where σx,σy,σz are the Pauli spin matrices, and the desired rotation, azimuth and polar angles θ, φ and α are achieved by setting the interaction time t, phase φ and de-

Rotary echoes

A basic means of reducing dephasing in Rabi flopping is the rotary echo [22], which may be considered the simplest composite rotation. Reminiscent of Hahn’s spin echo [27], this is a repeated application of the sequence θθθθθ; when θ = 360°, as illustrated in Figure 3, the 180° phase shift every whole Rabi cycle causes a periodic reflection of state vector trajectories and realignment, or echo, of divergent states. Figure 3 shows the remarkable reduction in Rabi flopping dephasing obtained with this technique, and the good agreement between experiment and simulation demonstrates the enduring coherence for individual atoms. Simulations for the measured velocity distribution and a Rabi frequency fRab = 200 kHz (where fRab is the pulse duration for optimal ensemble inversion) show flopping with an exponentially falling contrast with a time constant of about 250 µs, equivalent to 50 Rabi cycles. Experimentally, path length variations, and drifts in the beam intensities and single-photon detuning Δ, cause the fringe visibility to fall over

FIG. 3. Upper hyperfine state population |p⟩ as a function of Raman interaction time t: (a) regular Rabi flopping; (b) Rabi flopping with rotary echoes; (c) highly-sampled data for the indicated portion of (b). Open circles are experimental data; solid curves are numerical simulations for a double-Gaussian.

(d) 360°

(e) 360°, 360°, 360°, 360°, 360°, 360°
100 − 200 µs: the initial visibility reflects the residual Doppler sensitivity at our modest Rabi frequencies.

### Composite pulses

As rotary echoes are of limited use beyond revealing underlying coherence, our focus in this paper is upon composite pulses: sequences of rotations that together perform a desired manipulation of the state vector on the Bloch sphere with reduced dephasing from systematic inhomogeneities. Of the many sequences developed for NMR applications [13], we consider here just a few of interest for inversion in atom interferometry and such experiments. The sequences vary in two key respects.

First, it is common to distinguish between (a) general rotors, which are designed to apply the correct unitary rotation to any arbitrary initial state; and (b) point-to-point pulses, which work correctly only between certain initial and final states and which for other combinations can be worse than a simple π pulse. Some composite inversion pulses suitable for atom interferometry are summarized in Table I.

Secondly, each pulse sequence may be characterized by its sensitivity to variations in the interaction strength and tuning, which instead of the intended rotation propagator \(U(\theta, \phi, \alpha)\) result in the erroneous mapping \(V(\theta, \phi, \alpha)\). Pulse-length (or -strength) errors, associated with variations in the strength of the driving field or interaction with it, appear as a fractional deviation \(\epsilon = \Delta \theta / \theta\) from the desired rotation angle so that, for the example of a simple Rabi pulse, for small \(\epsilon\),

\[
V(\theta, 0, 0) = U((1 + \epsilon)\theta, 0, 0)
= U(\theta, 0, 0) - \epsilon \frac{\theta}{2} \left[ \sin \left( \frac{\theta}{2} \right) \mathbf{1} + i \cos \left( \frac{\theta}{2} \right) \sigma_x \right] + O(\epsilon^2). \tag{2}
\]

**Off-resonance** errors meanwhile correspond to tilts of the rotation axis due to offsets \(f = \delta / \Omega_{\text{eff}}\) in the driving field frequency, so that, for the same example and small \(f\),

\[
V(\theta, \phi, 0) = U \left( \theta, \phi, \sin^{-1}(f) \right)
= U(\theta, \phi, 0) + f i \sin \left( \frac{\theta}{2} \right) \sigma_x + O(f^2). \tag{3}
\]

It is common to describe the dependence upon \(\epsilon\) and \(f\) of the operation fidelity

\[
F = |\langle \psi | V^\dagger U | \psi \rangle|^2, \tag{4}
\]

which contains only even powers of \(\epsilon\) and \(f\). The leading-order uncorrected terms in the corresponding infidelity \(I = 1 - F\) are given in Table I for Rabi pulses in our system, pulse-length errors are caused by intensity inhomogeneities and mixed transition strengths, and off-resonance errors are due to Doppler shifts. Detunings such as Doppler shifts are also accompanied by pulse-length errors of order \(f^4\), and intensity variations similarly cause light shifts and thus small off-resonance errors.

We have compared the general rotor sequences known as CORPSE [14], BB1 [15] and KNILL [16], and the point-to-point WALTZ [17] sequence, designed for transfer between the poles of the Bloch sphere. The sequences last from three to five times longer than a Rabi π pulse, but all give higher fidelities and greater detuning tolerances. To characterise each inversion sequence experimentally, we measure the ensemble mean fidelity, equal to the normalized population \(|c_2|^2\) of state \(|2\rangle\). These are shown over a range of normalised laser detunings \(\delta / \Omega_{\text{eff}}\) for opposite circular Raman polarizations in Figure I and for orthogonal linear polarizations in Figure J. The displacement of the peak from \(\delta = 0\) shows the light shift in each case.

By truncating each sequence, we also determine the state population evolution, shown in Figure K for circular beam polarizations at the optimum Raman detuning. Experimental fidelities are all presented without correction for the beam overlap factor \(S\), described in the Methods section.

**Table I.** Common composite inversion pulses. The theoretical fidelity \(F\) depends upon the atom cloud temperature as shown in Figure L and is from simulations for typical parameters given in Table M. Bold values indicate best performance at \(\delta = 0\), which reflects the leading-order terms in the fidelity and their coefficients. PP: point-to-point, GR: general rotor.

| Composite Pulse | Type | Rotation Sequence \(\theta, \phi, \ldots\) | Leading order | total angle | \(F(\sigma^+ - \sigma^-)\) | \(F(\pi^+ - \pi^-)\) |
|-----------------|------|--------------------------------------|--------------|--------------|-----------------|-----------------|
| Rabi π-pulse    | PP   | 180\(\theta\)                       | \(\epsilon^2\) | \(f^2\)      | 180° 0.47       | 0.73            |
| CORPSE          | GR   | 60\(\theta\),300,180,420\(\theta\)  | \(\epsilon^2\) | \(f^4\)      | 780° 0.61       | 0.79            |
| KNILL           | GR   | 180\(2\theta\),180\(2\phi\),180\(2\alpha\) | \(\epsilon^4\) | \(f^4\)      | 900° 0.64       | **0.89**        |
| BB1             | GR   | 180\(\theta\),180\(3\theta\),180\(3\phi\) | \(\epsilon^6\) | \(f^2\)      | 900° 0.56       | 0.80            |
| 90-360-90       | PP   | 90\(\theta\),300,120,90\(\theta\)   | \(\epsilon^5\) | \(f^2\)      | 540° 0.59       | 0.82            |
| SCROFULOUS      | GR   | 180\(\theta\),180\(\phi\),180\(\alpha\) | \(\epsilon^6\) | \(f^2\)      | 540° 0.44       | 0.72            |
| LEVITT          | PP   | 90\(\theta\),180,90\(\theta\)       | \(\epsilon^2\) | \(f^2\)      | 360° 0.70       | 0.86            |
| 90-240-90       | GR   | 90\(\theta\),240,30,90,240\(\theta\) | \(\epsilon^2\) | \(f^2\)      | 420° 0.63       | 0.88            |
| 90-225-315      | PP   | 90\(\theta\),225,180,315\(\theta\)  | \(\epsilon^2\) | \(f^2\)      | 630° 0.71       | **0.89**        |
| WALTZ           | PP   | 90\(\theta\),180,270\(\theta\)      | \(\epsilon^2\) | \(f^2\)      | 540° **0.77**   | 0.88            |
FIG. 4. Measured upper state populations (circles) after various $\pi^+ - \pi^-$ inversion sequences, as functions of Raman detuning. Simulations (lines) are for a temperature, laser intensity and detuning found by fitting, within known uncertainties of measured values, to results for a basic $\pi$ (180°) pulse (a).

FIG. 5. Measured upper state populations (circles) after various $\sigma^+ - \sigma^+$ inversion sequences, as functions of Raman detuning. Simulations (lines) are for a temperature, laser intensity and detuning found by fitting, within known uncertainties of measured values, to results for a basic $\pi$ (180°) pulse (a).

DISCUSSION

Our experimental results and theoretical simulations demonstrate general characteristics of coherent manipulations, as well as the differences between different composite pulse sequences. In each case, the single-photon light shift due to the Raman beams is apparent in a detuning of the spectral peak from the low intensity resonance frequency, and features that are resolved in the case of $\pi^+ - \pi^-$ Raman polarizations, for which the light shift is independent of Zeeman sub-state, are blurred into a smooth curve for $\sigma^+ - \sigma^+$ polarizations. Temporal light shift variations as the pulse sections begin and end will distort the composite sequences, but appear to have little effect upon the overall performance. Spatial beam inhomogeneities, the sub-state-dependent Raman coupling strengths, and the Doppler shift distribution, should all to an extent be corrected by the composite pulses.

Our composite pulse sequences vary in the degree to which they cancel pulse length and off-resonance errors, with the CORPSE pulse suppressing only off-resonance effects, the BB1 tolerating only pulse length errors, and the KNILL pulse correcting the quadratic terms in both. Accordingly, the CORPSE sequence shows the greatest insensitivity to detuning, while the BB1 and KNILL pulses show higher peak fidelities. Although the BB1 is regarded as the most effective for combating pulse-length errors, we find that pulses that nominally correct for off-resonance effects only can provide greater enhancements in the peak fidelity and spectral width overall.

All three general rotors are out-performed in peak fidelity by the point-to-point WALTZ sequence, which has already been used for atom interferometer augmentation pulses [28]. This pulse is expected to enhance very small errors, but limit their effect to 5% for $|f| \lesssim 1.1$. In the $\sigma^+ - \sigma^+$ configuration, we observe that the WALTZ pulse nearly halves the infidelity $I$ upon which the interferometer contrast depends, from 0.58 for the basic $\pi$ pulse to 0.33. In the $\sigma^+ - \sigma^+$ configuration, the improvement is from $I = 0.35$ to $I = 0.24$, and the fidelity is maintained as predicted for detunings $|\delta_L| \lesssim \Omega_{\text{eff}}$; beyond this, it falls more gently so that, at $|\delta_L| \approx 3\Omega_{\text{eff}}$, it is over five times that for a $\pi$ pulse.

We note that, as the pulse durations in our experiments were chosen by optimizing the $\pi$ pulse fidelity, slight improvements might be possible for the other sequences, both because it is the atomic ensemble average that matters and because the bandwidths of our modulators cause
small distortions around the pulse transients.

The close agreement of our experimental results and theoretical simulations demonstrates both the validity of our simple model of the velocity and Zeeman state distributions and the durable underlying coherence of individual atomic states. In each case, the simulation parameters are based upon the measured laser intensities and detunings, which are adjusted within known uncertainties to match the results for a basic $\pi$ ($180^\circ$) pulse; the deduced values are listed in Table II. As the measured efficiencies depend upon experimental conditions that vary between data sets, we have simulated the performance of the $\pi$ pulse and KNILL and WALTZ sequences under consistent conditions, for a range of velocity distributions and for two different Zeeman sub-state distributions. Figure 7 demonstrates the expected decrease in fidelity with increasing atom cloud temperature and with the population of multiple Zeeman levels. For low temperatures ($\sigma_v < 5v_R$), the spectral width of the Rabi $\pi$ pulse at $\Omega_{\text{eff}} \sim 2\pi \times 350\,\text{kHz}$ exceeds the Doppler-broadened linewidth, and the peak fidelity is determined by the variation in Raman coupling strength between different Zeeman sub-states; the best fidelity is hence obtained with the superior pulse-length error performance of the KNILL sequence. For warmer samples, Doppler off-resonance errors dominate, and the CORPSE pulse is better. For atoms that are spin-polarized into a single Zeeman level, the performance of all pulses is improved, and the preference for the WALTZ pulse extends to slightly lower temperatures. Table III summarizes these results for conditions that are typical for our experiments, and also shows the theoretical performance of some other popular composite pulse sequences.

Our results show that the principal errors in the coherent manipulation of cold atoms are due to systematic inhomogeneities in the laser intensity, atomic velocity and Zeeman sub-state, and may hence be significantly reduced by composite pulse techniques. Near the recoil limit, we predict that instead of the the maximum $\pi$ pulse fidelity of 0.96 it should be possible to achieve fidelities in excess of 0.99, allowing many more augmentation pulses to impart a greater separation between the interferometer paths and hence an elevated interferometric sensitivity without losing atoms through spin squeezing. The greater tolerance of Doppler shifts similarly allows interferometry to be performed without further loss through velocity selection.

Atom interferometers require not only augmentation $\pi$ pulses, but beam-splitter/recombiner $\pi/2$ pulses ($90^\circ$), and for these it is likely that quite different composite pulse sequences will be required to minimize the effects of experimental variations upon the composition and phase of the quantum superposition: the solution depends upon the balance of different sources of error, and the relative importance of their different effects upon the final states. Cold atom interferometers are likely to differ in both respects from the NMR systems for which most established composite pulse sequences were developed. In our system, pulse length and off-resonance errors are not only conflated, they are to some extent correlated, for
the light shift is responsible for both.

The best solutions need not be those which optimize the fidelities of the individual interferometer operations, for it is likely that errors after the first beamsplitter, for example, could to some extent be corrected by the recombines. It is interesting to note that while, for an equal superposition initial state \( |\psi\rangle = \frac{1}{\sqrt{2}}(|g\rangle + e^{i\phi}|e\rangle) \) in

\[ |\psi\rangle = \frac{1}{\sqrt{2}}(|g\rangle + e^{i\phi}|e\rangle) \]

the presence of off-resonance errors, a single WALTZ inversion performs more poorly than a basic \( \pi \) pulse, a sequence of two WALTZ pulses gives an efficient \( 2\pi \) rotation, and that when two pairs of WALTZ pulses were applied as in the large area interferometer in [28], readout contrast was increased. The development of composite pulse sequences for atom interferometry should therefore consider the performance of the interferometer as a whole.

METHODS

Experimental setup

\(^{85}\)Rb atoms are initially trapped and cooled to \( \sim 250 \) \( \mu \)K in a standard 3D magneto-optical trap (MOT) to give about \( 2 \times 10^7 \) atoms in a cloud about 500 \( \mu \)m in diameter. The MOT magnetic fields are extinguished, the beam intensities ramped down, and the cloud left to thermalise in the 3D molasses for 6 ms, after which the temperature has fallen to \( \sim 50 \) \( \mu \)K. The velocity distribution exhibits a double-Gaussian shape, as shown in Figure 2, because atoms at the centre of the molasses undergo more sub-Doppler cooling than those at the edges [48]. The \( |5S_{1/2}, F = 2\rangle \rightarrow |5P_{3/2}, F = 3\rangle \) repumping beam is then extinguished, and the atoms are optically pumped for 300 \( \mu \)s into the \( |5S_{1/2}, F = 2\rangle \) ground hyperfine state by the cooling laser, which is detuned to the red of the \( |5S_{1/2}, F = 3\rangle \rightarrow |5P_{3/2}, F = 4\rangle \) transition. Three mutually orthogonal pairs of shim coils cancel the residual magnetic field at the cloud position, and are calibrated by minimising the spectral width of a Zeeman-split, velocity-insensitive (co-propagating) Raman transition. From the spectral purity of the measured velocity distribution, we deduce the residual magnetic field to be less than 10 mG, equivalent to a Zeeman splitting of \( \sim 5\) kHz in \( m_F \), and hence that the Zeeman sub-levels for each hyperfine state are degenerate to within a fraction of the typical Rabi frequency \( \Omega_{\text{eff}} > 200 \) kHz. After preparation, we apply the Raman pulses to couple the states \( |5S_{1/2}, F = 2\rangle \) and \( |5S_{1/2}, F = 3\rangle \), and then measure the resultant \( |5S_{1/2}, F = 3\rangle \) state population by detecting fluorescence when pumped by the cooling laser.

The apparatus used to generate our Raman pulses is shown schematically in Figure 3. The beams are generated by spatially and spectrally splitting the continuous-wave beam from a 780 nm external cavity diode laser, red-detuned from single-photon resonance by \( \Delta \approx 2\pi \times 10 \) GHz. The beam is spatially divided by a 310 MHz acousto-optical modulator (AOM), and the remainder of the microwave frequency shift is generated by pass-

ing the undeflected beam from the AOM through a 2.726 GHz electro-optical modulator (EOM). We modulate the EOM phase and frequency using an in-phase and quadrature-phase (IQ) modulator, fed from a pair of arbitrary waveform generators. The carrier wave at the output of the EOM is removed using a polarising beamsplitter cube [49], and temperature-dependent birefringence within the EOM is countered by active feedback to a liquid crystal phase retarder [50]. The remaining off-resonant sideband is removed using a stabilized fibre-optic Mach–Zehnder interferometer [51].

Following pre-amplification of the EOM sideband by injection-locking a c.w. diode laser, the two spatially separate, spectrally pure Raman beams are then individually amplified by tapered laser diodes, recombined with orthogonal polarisations and passed through an AOM, whose first-order output forms the Raman pulse beam. Before they are separated at a polarising beam-splitter, a Pockels cell allows the beam polarisations to be switched and hence their propagation directions exchanged. The AOM rise and fall times alter the effective pulse timing but are not included in the sequence design; proper compensation could further improve the observed fidelity.

Each beam is passed through a Topag GTH-4-2.2 refractive beam shaper and 750 mm focal length lens to produce an approximately square, uniform beam whose intensity varies by only 13% across the extent of the MOT cloud. The beams measure \( \sim 2 \) mm square and each has

FIG. 8. Schematic of the experimental setup of the Raman beams: ECDL – external-cavity diode laser; PBSC – polarising beamsplitter cube; TA – tapered amplifier; PC – Pockels cell; OSA – optical spectrum analyser; BSh – beam shaper and focussing lens. The annotation bubbles show sketches of the beam spectrum at each preparation stage. For clarity, the injection-locked pre-amplifier following the EOM is omitted.
an optical power of 50 mW, corresponding to an intensity of \( \sim 1.3 \text{ W cm}^{-2} \). Compared with the large-waist Gaussian beams required for the same spatial homogeneity, this provides a significantly higher intensity and as a result our system exhibits two-photon Rabi frequencies of \( \Omega_{\text{eff}} \approx 2\pi \times 250 \text{ kHz} \). Using a shorter focal-length lens in the beam path to produce a smaller top-hat, we have observed a higher Rabi frequency of \( \Omega_{\text{eff}} \approx 2\pi \times 500 \text{ kHz} \), but are then limited by the number of atoms that remain within the beam cross-section. Although the phase profile of the top-hat beam is non-uniform [52], we calculate that an individual atom will not traverse a significant phase gradient during a few-\( \mu \text{s} \) pulse sequence.

Because our Raman beams illuminate a smaller region than the cooling and repump beams used to determine the final state population, a fraction of the expanding atom cloud contributes to the normalization signal without experiencing the Raman pulse sequence. We have characterized the time dependence of this effect, and scale our simulated upper state populations in Figures 3 and 4) we ran through different values of the detuning in a composite pulses immediately after taking data on its

\[ E = E_1 e^{i(k_1 \cdot x + \omega_{L1} t + \phi_1)} + E_2 e^{i(k_2 \cdot x + \omega_{L2} t + \phi_2)}, \]  

where on resonance \( \omega_{L1} = \omega_1 - \omega_1 \) and \( \omega_{L2} = \omega_3 - \omega_2 \), \( E_{1,2} \) are the Raman beam amplitudes, and we define \( \phi_1 = \phi_2 \) as the effective phase of the Raman field. For the analytical solutions to the time-dependent Schrödinger equation for this system in the interaction picture, we refer the reader to [55].

The effective Rabi frequency of the Raman transition (\( \Omega'_{\text{eff}} \)) depends on (a) the respective Clebsch-Gordan coefficients of the Raman route whose relative amplitudes are shown in Figure 1 (b) the intensity of the driving field, which in our simulations is taken to be temporally ‘square’ and (except in Figure 3) spatially homogeneous, and (c) the detuning of the driving field from the atomic resonance, which is Doppler-shifted by the atom’s motion. It follows that for a Doppler-broadened ensemble of atoms distributed across degenerate sublevels, we expect a distribution of \( \Omega'_{\text{eff}} \) values and therefore a dephasing of atomic states during a Raman pulse. Consequently, the \( \pi \) pulse efficiency will be unavoidably limited to much less than unity in the absence of effective error correction.

To simulate the system we numerically calculate the hyperfine state amplitudes \( c_{1,2,3}(p) \) for a period \( t \) of interaction with the Raman beams, and integrate over all Raman routes and velocity classes. The atoms are taken initially to be evenly distributed across the Zeeman \( m_F \) sub-levels of \( [5S_{1/2}, F = 2] \), and opposite-circularly polarised Raman beams are considered to drive \( \sigma^+ \) dipole-allowed transitions via the Raman routes shown in Figure 1 where, regardless of the quantisation axis, conservation of angular momentum requires that \( \Delta m_F = 0 \).

The primary free parameters in our simulations are the velocity distribution, which we model as having two Gaussian components similar to those fitted to the measured distribution in Figure 2 the sampling factor \( S \) and the laser intensity \( J \). To account for experimental variations, we allow small adjustments from measured values to give a closer fit to the data; the values used for our various simulations are listed in Table II.

Theoretical model

We drive two-photon stimulated Raman transitions between the two hyperfine ground states in \(^{85}\text{Rb} \), as shown in Figure 1. When the two Raman beams with angular frequencies \( \omega_{L1,2} \) and wavevectors \( k_{1,2} \) travel in opposite directions (\( k_1 \approx - k_2 \)), the Raman interaction is velocity-sensitive and each transition is accompanied by a two-photon recoil of the atom \( \hbar \omega \equiv k_{1,2}^2 / 2m \approx \pm 2k_{1,2} \) as a photon is scattered from one Raman beam to the other. The internal state of the atom is therefore mapped to its quantised external momentum state. If an atom is prepared in the lower hyperfine state \( [5S_{1/2}, F = 2] \), which we label \( |1\rangle \), and the Raman transition couples this, via an intermediate virtual state \( |3\rangle \), to the upper hyperfine state \( [5S_{1/2}, F = 3] \), labelled \( |2\rangle \), then the momentum-inclusive basis in which we work is \( |1, p \rangle, |2, p + h\mathbf{k}_{\text{eff}} \rangle \).

For clarity, we henceforth omit the momenta and leave these implicit in our notation.

The Hamiltonian for the Raman system is [53]

\[ \hat{H} = \frac{\hat{p}^2}{2m} + \hbar \omega_1 |1\rangle \langle 1| + \hbar \omega_2 |2\rangle \langle 2| + \hbar \omega_3 |3\rangle \langle 3| - \mathbf{d} \cdot \mathbf{E}. \quad (5) \]

where \( \mathbf{p} \) is the momentum operator and differs in this equation from the ground state momentum \( \mathbf{p} \) principally through the introduction of the small Doppler shift to the resonance frequency due to the impulse imparted by the transition. The initial and final electronic states are taken to have energies \( \hbar \omega_1 \) and \( \hbar \omega_2 \), \( \mathbf{d} \) is the Raman electric dipole operator, acting via all intermediate states \( |j\rangle \), and the electric field of the two Raman beams counter-propagating along the \( z \) axis is given by

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of the single photon recoil velocity $\sigma$ and temporally invariant. The velocity distribution is taken to have two Gaussian components with widths populated, and the Raman beam intensities — which may differ because of different beam widths — are assumed to be spatially

| Table II. Parameters used in theoretical simulations. In each case, the hyperfine state $m_T$ to a temperature of $T = m \sigma^2/k_B = 1.48(\sigma_{\pi}/v_{\text{rec}})^2 \mu K$. † Also represents $\pi^+ - \pi^-$ as the B-field is set to compensate for Zeeman-like light shift. ‡ For the single sub-state $m_F = 0$, $\Omega_{\text{eff}} = 316 \times 2 \pi$ Hz, $t_s = 1.43 \mu s$. |

| Polarization $I_1$ $I_2$ $\Delta$ $B_x$ $\sigma_1$ $\sigma_2$ $a_{1/2}$ $S$ $\Omega_{\text{eff}}$ $t_s$ |
|---|---|---|---|---|---|---|---|---|---|---|
| $\sigma^+ - \sigma^+$ | 12.1 | 12.1 | 12.3 | 5 | 22.5 | 4 | 250 | 2 |
| $\sigma^+ - \sigma^+$ | 3 | 4.6 | 10 | -11 | 1.8 | 7.5 | 4 | 4.5 | 110 |
| $\sigma^+ - \sigma^-$ | 12 | 17 | 15 | -101 | 3 | 10 | 3 | 0.95 | 200 | 2.5 |
| $\sigma^+ - \sigma^+$ | 14 | 21 | 9.0 | -11 | 2.5 | 9 | 2 | 0.9 | 357 | 1.4 |
| $\pi^+ - \pi^-$ | 14 | 21 | 8.0 | -11 | 2.5 | 9 | 2 | 0.9 | 417 | 1.2 |
| $\sigma^+ - \sigma^-$ | 14 | 21 | 8.5 | -11 | 2.5 | 9 | 2 | 0.9 | 385 | 1.3 |
| $\sigma^+ - \sigma^+$ † | 14 | 21 | 9 | -261 | - | various | 0 | 1 | 350† | 1.58$^{\dagger}$ |

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