1 Theory for Quench Dynamics in a Harper Ladder

1.1 Bloch Bands and Wannier Functions of the Harper Ladder

In this section we consider the non-interacting limit where $U = 0$. Despite the presence of the magnetic flux $\Phi$ per plaquette, a gauge choice can be made for the ladder geometry with a two-site unit cell corresponding to an individual rung. Now we derive the cell-periodic Bloch wavefunctions $|u_{\pm}(q)|$ of the resulting two bands. After deriving their dispersion relations $\epsilon_{\pm}(q)$, we discuss particle-hole symmetry and calculate the corresponding Wannier functions $w_{\pm}(x, y)$.

1.1.1 Bloch Hamiltonian

To derive the Bloch Hamiltonian, we make a gauge choice where the tunneling elements describing hoppings in the positive $x$-direction along the lower (upper) leg of the ladder have a complex phase $\Phi/2$ ($-\Phi/2$). The resulting unit-cell is defined by the two sites of a rung of the ladder, with the two states $|U\rangle, |D\rangle$ corresponding to the upper (“up”) and lower (“down”) leg of the ladder. Notably, the unit-cell is not related to the magnetic unit-cell which encloses an integer number of magnetic flux quanta, reflecting the one-dimensional character of the ladder system.

Without the rung coupling $J$, the Bloch Hamiltonians of the two decoupled legs are given by $-2K\cos(q \pm \Phi/2)$ where the phases $\pm\Phi/2$ only lead to a shift of the two quasimomenta $q$. In terms of Pauli matrices $\hat{\sigma}^{x,z}$, defined in the basis $|U\rangle, |D\rangle$ by the relations $\hat{\sigma}^{x}|U\rangle = |U\rangle$ and $\hat{\sigma}^{z}|D\rangle = -|D\rangle$, this Bloch Hamiltonian can be written $\hat{H}(q) = -K(\nu_+(q) + \nu_-(q)\hat{\sigma}^{z})$, where

$$\nu_\pm(q) = \cos(q + \Phi/2) \pm \cos(q - \Phi/2).$$

The coupling $J$ between the two legs is described by an additional term $-J\hat{\sigma}^{x}$. The final Bloch Hamiltonian of the ladder system in the presence of the artificial gauge field, for which $\hat{H}(q)|u_{\pm}(q)\rangle = \epsilon_{\pm}(q)|u_{\pm}(q)\rangle$, is thus given by

$$\hat{H}(q) = -K\nu_+(q) - (J\hat{\sigma}^{x} + K\nu_-(q)\hat{\sigma}^{z}).$$

The cell-periodic Bloch functions and the corresponding energies can easily be calculated,

$$|u_{+}(q)\rangle = \cos\frac{\vartheta_q}{2}|U\rangle + \sin\frac{\vartheta_q}{2}|D\rangle,$$

$$|u_{-}(q)\rangle = \cos\frac{\vartheta_q}{2}|D\rangle - \sin\frac{\vartheta_q}{2}|U\rangle,$$

where the angle $\vartheta_q$ is defined by

$$J\hat{e}_x + K\nu_-(q)\hat{e}_z = \sqrt{J^2 + K^2\nu_2^2(q)}(\sin\vartheta_q\hat{e}_x + \cos\vartheta_q\hat{e}_z).$$

The dispersion relations of the two bands are given by

$$\epsilon_{\pm}(q) = -K\nu_+(q) \mp \sqrt{J^2 + K^2\nu_2^2(q)}.$$

These dispersion relations are shown in Fig. 3c in the main part of the paper.

Physically the two bands ($\pm$) can be understood as hybrids of two counter-propagating chiral edge states. Switching between the two kinds of hybridization, which are symmetric (+) and antisymmetric (−) superpositions of the upper and lower legs of the ladder respectively, can be formalized by the particle-hole operator which we discuss next.

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1.1.2 Wannier functions

The Wannier functions $w_\tau$ corresponding to the two bands ($\tau = \pm$) are defined by

$$w_\tau^\mu(x - x_j) = \frac{1}{\sqrt{2\pi}} \int_{-\pi}^{\pi} dq \ e^{iq(x-x_j)} u_\tau^\mu(q),$$

(S3)

where $x, x_j \in \mathbb{Z}$ label one-dimensional lattice sites along the ladder and $\mu = U, D$ denotes the two legs of the ladder. Note that $x$ and $\mu$ represent the coordinates of the Wannier function $w_\tau$ corresponding to the band labeled by $\tau$; similarly $u_\tau^\mu(q)$ labels the component corresponding to state $|\mu\rangle$ of the Bloch wavefunction $|u_\tau(q)\rangle$.

**Numerical results:** The Fourier transformation Eq. (S3) can be done numerically, and we find for all values of $\Phi$ that the Wannier functions are very well localized on a single rung of the ladder. For $\Phi \neq 0$ there is a small amplitude on the neighboring rungs, but even for the largest values of $\Phi$ less than 10% of the orbit leaks onto neighboring rungs.

As an example, we show the Wannier functions $w_\tau^U(x)$ and $w_\tau^D(x)$ for the case $\Phi = \pi/2$ in Figs. S1a and S1b. To a rather good approximation they are given by

$$|w_\pm(x - x_i)\rangle \approx \delta_{x,x_i} \langle |D\rangle \pm |U\rangle \rangle / \sqrt{2},$$

i.e. in terms of bosonic operators:

$$\hat{a}^\dagger_{i,\pm} \approx \left( \hat{a}^\dagger_{i,D} \pm \hat{a}^\dagger_{i,U} \right) / \sqrt{2}.$$ 

While this expression is exact in the absence of the gauge field, $\Phi = 0$, the approximation is not sufficient to understand the chirality in the system, which arises from the coupling of the motion along and transverse to the ladder.

![Figure S1](image.png)

**Figure S1:** a. The real and imaginary parts of the Wannier function $w_\tau^\mu(x)$, for $\mu = U, D$, is shown, for $J = 34.1$ Hz $K = 11.4$ Hz, and $\Phi = \pi/2$. b. The real and imaginary parts of the Wannier function $w_\tau^D(x)$, for $\mu = U, D$, is shown, for $J = 2K$ and $\Phi = \pi/2$.

**General properties:** From Eq. (S3) a few general properties of the Wannier functions can be derived exactly using the symmetries of the system. The first concerns the relation between $|w_+\rangle$ and $|w_-\rangle$, which follows from the particle-hole symmetry of the system,

$$w_\tau^U(x) = \tau \ w_\tau^D(x), \quad \tau = \pm 1.$$ 

Second we discuss how the values of the Wannier functions on different legs are related. Using that the system is invariant under simultaneous exchange of the upper and lower legs, $U \leftrightarrow D$, and inversion of the flux, $\Phi \rightarrow -\Phi$, we can show that

$$w_\tau^U(x) = \tau \ (w_\tau^D)^* = -(w_\tau^U(x))^*.$$
Finally we can also derive the relations of Wannier functions at $\pm x$. Because the system is invariant under simultaneous spatial inversion and exchange of the upper and lower legs, it follows that

$$w^U_r(-x) = \tau w^D_r(x), \quad w^D_r(-x) = \tau w^U_r(x).$$

1.2 Quench Dynamics of the Interacting System – Physical Picture

To understand the emergence of chirality in the quantum walk of two interacting bosons, we calculate numerically the decomposition of the initial state $|\psi_{\text{initial}}\rangle$ into the eigenstates $|\phi_n\rangle$ of the system,

$$|\psi_{\text{initial}}\rangle = \sum_n c_n |\phi_n\rangle.$$  

In Fig. 5 (c) of the main text, we first show the squared amplitudes $|c_n|^2$ plotted against the corresponding eigenenergies $E_n$ as a function of the flux $\Phi$. For better visualization we introduced bins, each of which contains about 10 neighboring eigenenergies, and summed up all the overlaps $|c_n|^2$ corresponding to each bin. Because the squared amplitudes $|c_n|^2$ are conserved in the dynamics, this decomposition allows to understand the behavior of the system at long times.

To understand the physics of the eigenstates $|\phi_n\rangle$ themselves, we proceed by calculating their decomposition into free two-particle eigenstates,

$$|\phi_n\rangle = \sum_{k_1,k_2} \left[ \phi_{n,+}^{k_1,k_2}(k_1,k_2) \hat{a}_{k_1,+} \hat{a}_{k_2,+}^{\dagger} + \sqrt{2} \phi_{n,-}^{k_1,k_2}(k_1,k_2) \hat{a}_{k_1,-}^{\dagger} \hat{a}_{k_2,-}^{\dagger} / \sqrt{2} \phi_{n,-}^{k_1,k_2}(k_1,k_2) \hat{a}_{k_1,-}^{\dagger} \hat{a}_{k_2,-}^{\dagger} / \sqrt{2} \phi_{n,-}^{k_1,k_2}(k_1,k_2) \hat{a}_{k_1,-}^{\dagger} \hat{a}_{k_2,-}^{\dagger} \right] |\text{vac}\rangle,$$

where the two-boson wavefunction $\phi_{n,+}^{k_1,k_2}(k_1,k_2) = \phi_{n,-}^{k_2,k_1}(k_2,k_1)$ is symmetric (and analogously for $\phi_{n,-}^{k_1,k_2}$). Here the operator $\hat{a}_{k,\tau}^{\dagger}$ creates a boson at quasimomentum $k$ in the band labeled by $\tau = \pm$.

![Figure S2](image_url)

**Figure S2: Decomposition of interacting into free two-particle eigenstates:** a. The overlap $p_{++}(n)$ is shown (color code) as a function of the eigenenergies $E_n$ and the flux $\Phi$ per plaquette of the synthetic gauge field, see Eq. (S5). In b and c, the calculation is repeated for $p_{+-}$ and $p_{-+}$, respectively.

To study the influence of interactions on the free eigenstates $\hat{a}_{k_1,\tau_1}^{\dagger} \hat{a}_{k_2,\tau_2}^{\dagger} |0\rangle$, we calculate their overlaps with the eigenstates $|\phi_n\rangle$ of the interacting system. In Fig. S2 (a) the amplitudes summed over all quasimomenta $k_1, k_2$ are shown for $\tau_1 = \tau_2 = +$,

$$p_{++}(n) = \sum_{k_1,k_2} |\phi_{n,+}^{k_1,k_2}(k_1,k_2)|^2,$$

again as a function of the flux per plaquette $\Phi$ of the synthetic gauge field and the eigenenergy. We used the same binning method for the energies as described for the overlaps $|c_n|^2$ above. In Fig. S2 (b), (c) we repeat this calculation for $p_{+-}(n)$ and $p_{-+}(n)$ defined using $|\phi_{n,-}^-|^2$ and $|\phi_{n,-}^-|^2$, respectively.

From these figures we draw the conclusion that the energy spectrum of the interacting Hamiltonian consists of two main features. Firstly, we recognize three broad bands of eigenstates (labeled $++$, $+-$, and $-+$) which are very similar to the states of free bosons at $U = 0$. Most of these states are scattering states, i.e. the two bosons are likely to be far away from each other and their wavefunction can be well approximated.
by two independent plain waves in that case, $\hat{a}^\dagger_{k_1,\tau_1} \hat{a}_{k_2,\tau_2}|0\rangle$. This justifies labeling these states by the two quantum numbers $|\tau_1,\tau_2\rangle$ as done in the main text of the paper. In addition to the scattering states, we find three narrow bands of bound states in Fig. S2 (a) - (c) corresponding to repulsively bound pairs with a large effective mass. The energetically lowest branch mostly consists of particles in the lowest (+) band. The two upper branches, with energy $\sim U$, mostly correspond to one boson in each band and two bosons in the $-$ band, respectively.

Figure S3: **Chirality of Bloch bands:** The dispersion relations $\epsilon_\pm(q)$ of the two Bloch bands (lower, +, and upper, $-$) are plotted, for $J = 2\pi \times 34.1\ Hz$ and $K = 2\pi \times 11.4\ Hz$ close to the experimental values. The color code indicates the chirality $C^\pm(q)$ of the two bands, determined by Eq. (S6). Different values of $\Phi$ are considered: a, $0.11 \times 2\pi$, b, $0.3 \times 2\pi$, c, $0.4 \times 2\pi$, and d, $0.5 \times 2\pi$.

Finally, we describe how we analyzed the chirality of the interacting eigenstates $|\phi_n\rangle$, shown in Fig. 5 (c) in the main text. To this end, we first calculated the chirality of the single-particle eigenstates. For a given quasimomentum $k$ the chirality of band $\tau$ can be defined by

$$C^\tau(k) = \text{sign}(v_g^\tau(k)) \times (p_{gU}^\tau(k) - p_{gD}^\tau(k)),$$

where $\text{sign}(v_g^\tau(k))$ denotes the sign of the group velocity $v_g^\tau(k) = \partial_k \omega_k^\tau$, and $p_{gU,D}^\tau(k) = |u_{gU,D}^\tau(k)|^2$ are the probability amplitudes for a particle to reside on the upper (U) or lower (D) leg of the ladder. As an example, this chirality is shown color-coded in Fig. S3 for the experimentally relevant bandstructures.

To obtain the chirality of the interacting eigenstates $|\phi_n\rangle$, we combine the projection to free two-particle states with the single-particle chirality. This allows us to define the chirality of $|\phi_n\rangle$ as

$$C_n = \sum_{k_1,k_2} \left[ |\phi_n^{++}(k_1,k_2)|^2 \left(C^+(k_1) + C^-(k_2)\right) + |\phi_n^{+-}(k_1,k_2)|^2 \left(C^+(k_1) + C^-(k_2)\right) + |\phi_n^{-+}(k_1,k_2)|^2 \left(C^+(k_1) + C^-(k_2)\right) \right].$$

Figure S4: Illustration of the leading-order terms contributing to the short-time perturbative expansion: The $O(i\hat{t}_R^2)$ paths interfere with the $O(i\hat{t}_R^3)$ path including on-site interactions on the upper leg of the ladder. An equivalent set of interfering paths, with the upper and lower legs exchanged, leads to the leading-
order non-vanishing chiral signal scaling like $Ut^5 \sin \Phi$, as described in the text.

In Fig. 5(d) of the main text we show $C_n$ as a function of the eigenenergies and the synthetic magnetic field, using the binning method described above.

### 1.3 Short-Time Expansion

As mentioned in the text, we compute $\langle \hat{n}_{i,1} - \hat{n}_{i,0} \rangle$ for the initial state $|\psi(t)\rangle = a_{0,1}^\dagger a_{0,0}^\dagger |\text{vac}\rangle$, where $i = 0$ is the center of the ladder. Using the Hamiltonian in Eq. (1) and $|\psi(t)\rangle = e^{-iHt/\hbar} a_{0,1}^\dagger a_{0,0}^\dagger |\text{vac}\rangle$, we Taylor expand in $t$ (valid for $t \ll \frac{\hbar}{\text{max}(J,K,U)}$) and compute $\langle \hat{n}_{i,1} - \hat{n}_{i,0} \rangle$. As expected, when $i = 0$, $\langle \hat{n}_{0,1} - \hat{n}_{0,0} \rangle = 0$. Then, for $i = 1$, the relevant non-vanishing terms in $|\psi(t)\rangle$ are

$$
|\psi(t)\rangle = \ldots - \frac{1}{2!} \left( \frac{t}{\hbar} \right)^2 (2JK((e^{-i\Phi/2} + \cos(\Phi/2))a_{1,0}^\dagger a_{0,0}^\dagger + (e^{i\Phi/2} + \cos(\Phi/2))a_{1,1}^\dagger a_{1,0}^\dagger)) |\text{vac}\rangle
$$

$$
+ \frac{1}{3!} i \left( \frac{t}{\hbar} \right)^3 (2JKU(e^{i\Phi/2}a_{1,1}^\dagger a_{1,0}^\dagger + e^{-i\Phi/2}a_{1,0}^\dagger a_{1,0}^\dagger)) |\text{vac}\rangle, \tag{S8}
$$

as illustrated in Figure S4. This yields ($i = -1$ is similar)

$$
\langle \hat{n}_{1,1} - \hat{n}_{1,0} \rangle = - \langle \hat{n}_{-1,1} - \hat{n}_{-1,0} \rangle = t^5 \left( \frac{U}{\hbar} \right) \left( \frac{KJ}{\hbar^2} \right)^2 \sin(\Phi). \tag{S9}
$$

The odd dependence on the flux, $\Phi$, arising from the on-site interaction occurring on the top versus the bottom leg, indicates that the effect is chiral. For small $t$, Figure S5 shows good agreement between equation S9 and numerical results, convincingly showing a $t^5$ dependence. In the $U \to \infty$ limit, we recompute the short-time expansion with hard-core bosons, and notice the non-vanishing terms in equation S8 require the asymmetry of the particles interacting on the top versus the bottom legs of the ladder. Such a case is not possible in the hard-core boson limit, and up to order $t^5$, all terms vanish.

![Figure S5: Log plot of both the analytical theory (AT) result $\langle \hat{n}_{1,1} - \hat{n}_{1,0} \rangle = t^5 \left( \frac{U}{\hbar} \right) \left( \frac{KJ}{\hbar^2} \right)^2 \sin(\Phi)$, which can be obtained using a short-time expansion, and the numerical results obtained from exact diagonalization (ED) for different flux values $\Phi$. Note the strong agreement for $t \lesssim 1 \text{ ms}$ at all flux and the clear $t^5$ dependence.](image-url)
2 Calibrations

2.1 Tunneling Calibration

We calibrate the tunneling rates in the presence of the Raman lattice and the overall lattice tilt. The tilt is engineered by applying a physical magnetic field gradient along the leg dimension of the ladder. We first find the frequency of the Raman lattice that restores tunneling along this leg dimension, which is also referred to as the \( x \) dimension throughout this text. As discussed in the main text, the Raman lattice is created by projecting a pair of beams with relative detuning \( \Delta \omega = E \) through our microscope objective, where \( E \) is the energy offset between neighboring lattice sites that arises from the applied lattice tilt. Experimentally, we find \( \Delta \omega \) by initializing a single atom in a one-dimensional lattice and observing the occupation of the original site as a function of detuning of the two beams. The obtained same-site occupation after a short time \( t = 8.5 \text{ ms} \ll \hbar/K \) is shown in Fig. S6. The detuning where the site occupation is minimal corresponds to the point where resonant tunneling is restored. For all experiments we set \( \Delta \omega/2\pi \approx 870 \text{ Hz} \), which is the minimum of a Gaussian fit to the data.

![Figure S6: Resonant frequency calibration](image)

Figure S6: **Resonant frequency calibration**: Occupation of the initial site in a tilted lattice after \( t = 8.5 \text{ ms} \) of evolution plotted against the detuning of the Raman beams.

The tunneling \( J \) along the rung is determined from a double-well oscillation. We initialize an atom on one side of a double-well potential and measure its time evolution. The occupation number undergoes oscillations between the sites with frequency \( 2J \). We obtain the tunneling rate \( J/h = 34 \text{ Hz} \) from a fit to the data. In order to characterize the tunneling rate \( K \) along the leg, we perform a single-particle quantum walk [1]. A single atom is initialized in a one-dimensional lattice along \( x \) and the evolution of the density distribution is measured (Fig. S7 (a)). For a quantum walk, we expect the density distribution on site \( i \) to evolve in time as

\[
\rho_i(t) = \left| \mathcal{J}_i \left( \frac{2}{\pi} \frac{K}{\delta} \sin(\pi \delta t) \right) \right|^2 ,
\]

where \( \mathcal{J}_i \) is the Bessel function of the first kind on site \( i \) and \( \delta \) is the residual lattice tilt (equivalent to residual Raman lattice detuning) [2]. The theoretical density distribution is fitted to the data (Fig. S7 (a)), yielding \( K/h = 11.4 \text{ Hz} \). We also investigate the dependence of the tunneling rates on the power of the Raman beams by performing single-particle quantum walks along \( x \) and \( y \) for several Raman lattice depths. The resulting tunneling rates and fits of the expected dependence are shown in Fig. S7 [3].
Figure S7: **Restored and suppressed tunneling calibration:** 

**a,** Quantum walk in a one-dimensional lattice along the leg dimension \(x\). 

**b,** Dependence of the tunneling rates along \(x\) (yellow) and \(y\) (blue) on the gauge field power. Here, \(J_{0,1}\) are Bessel functions of the first kind. They are scaled by the bare tunneling strengths 40 Hz along \(x\) and 50 Hz along \(y\) that we measure in the absence of tilt and Raman lattice.

### 2.2 Interaction and Tilt Calibration

We use photon-assisted tunneling to calibrate both the on-site interaction \(U\) and the lattice tilt \(E\) [4]. We start with a unity-filling Mott insulator. By modulating the optical lattice potential, we observe photon-assisted tunneling between neighboring sites at the modulation frequencies \(U \pm E\). From this, we extract \(U/h = 131\) Hz after scaling to the lattice depth that was used in the experiment. In addition, we independently calibrate the dependence of the tilt \(E\) on the voltage applied to the gradient coil.

### 2.3 Flux Calibration

The flux per plaquette depends on the wavevectors, \(k_1\) and \(k_2\), of the Raman lattice that drives the laser-assisted tunneling [5, 6]. The phase associated with each complex tunneling element is given by the phase of the Raman lattice at the origin (destination) site for rightward (leftward) tunneling terms. The phase of the Raman lattice at site \((m, n)\) is \(\phi_{m,n} = \delta k \cdot R_{m,n} = m\phi_x + n\phi_y\) where \(\phi_x = \delta k_x a\) and \(\phi_y = \delta k_y a\). The net phase for traversing a closed contour on the lattice gives \(\Phi = \phi_y\) phase per lattice site, which is independent of \(\phi_x\). For the experiments described in this paper, \(\phi_x = \pi\) to minimize the intensity of light fields needed to attain a given laser-assisted tunneling strength. The uniqueness of the presented apparatus lies in the tunability of the Raman lattice’s wavevector, which in turn leads to tunability of the flux per plaquette \(\Phi\).

The Raman lattice is produced by the interference of two blue-detuned optical beams that are projected through the same objective as the beams that produce the bare optical lattice. The wavevectors, \(k_1\) and \(k_2\), of the Raman lattice are tuned by two piezo mirrors, allowing for independent control of the angle of the beams in the image plane. By independently changing the wavevectors \(k_1\) and \(k_2\), the \(\delta k\), and hence the flux \(\Phi\), is dynamically tunable within a single experimental sequence. This method for tuning the flux does not require changing the wavelength of the Raman beams.
Figure S8: **Tuning flux:** From left to right, the three columns show an image of the light fields in the Fourier plane (FP), a schematic visualizing the lattice vectors related to the Raman beams, as well as the voltages applied to the piezo mirrors to achieve the given configuration of these beams, both for $\Phi = 0.55\pi$ (a) and $\Phi = 0$ (b). The Fourier plane images are color coded as follows: The gray beams are the 2D lattice beams used to create the static optical lattice, the brown beams create the Raman lattice, and diffuse gray light represents unintended reflections off other optical surfaces.

### 3 Experimental Sequence

All of the experiments described in this letter start with a 2-D, single layer Mott insulator of $^{87}\text{Rb}$ in a deep optical lattice ($V_x = V_y = 45E_r$, $E_r/2\pi \sim 1.24$ kHz) with a 680 nm spacing [7]. The experimental sequences for both the single-particle and interacting two-particle experiments are illustrated in Fig. S9.

#### 3.1 Initial State Preparation

##### 3.1.1 Preparation of the Initial Fock State Distribution

For both experiments we deterministically prepare an initial state from the $N = 1$ shell of a Mott insulator. We choose the number of atoms for each experiment by projecting an additional confining (or “cutting”) optical potential from a digital micromirror device (DMD) located in the Fourier plane of our imaging system [8]. This “cutting” potential is either a single-well or a double-well along one dimension, and a smoothed flattop potential along the other dimension. The additional potential is superimposed on top of the atoms which are still situated in a deep lattice. The atoms outside of the “cutting” potential are then removed from the system by turning off the optical lattice and applying an anti-confining potential to efficiently expel them from the system. The lattice is then ramped back on and the anti-confinement potential is turned off. As illustrated in Fig. S10, this sequence is first applied in the $x$- and then in the $y$-direction of the lattice such that either a $1 \times 1$ or a $2 \times 1$ initial state is produced. The loading efficiency of these states is $\approx 93\%$ and is largely dominated by the initial Mott insulator fidelity.

##### 3.1.2 Single-Particle State Preparation

There is an additional Landau-Zener preparation step for the single-particle experiments, which require an individual atom to be delocalized across the central rung of the ladder system. First, the ladder-forming double-well potential is projected on top of a deep, non-tilted lattice with a single atom being located on one side of the central double-well. While the tunneling is still suppressed, we add an additional tilt $\Delta \gg J$ with a physical magnetic field gradient along the rung direction, which is used to prepare the occupied site as the ground state of the tilted double-well system (cf. Fig. S10 (c)). Tunneling is then rapidly increased by ramping down the lattice potential (final parameters: $V_y = 11E_r$, $\Delta \approx 20$). Finally, we adiabatically prepare the $\Delta = 0$ ground state of the balanced double-well system by ramping down the magnetic field gradient.
Figure S9: **Experimental sequence**: Schematic showing the approximate ramps and relative timing of the $x$-, $y$-lattices, the $x$-, $y$-DMD-potentials, the $x$-, $y$-tilts, and the Raman lattice. The profiles of the DMD potentials are sketched for the dimension labeled on the left. The other dimension of the profiles is well described by a smooth flattop potential within the region used for the experiment. All optical ramps are changed exponentially in depth as a function of time and are sketched with a logarithmic y-scale here. The $x$-, $y$-tilts that are created by magnetic field gradients are, however, plotted and ramped linearly.
over a period of 100ms, which results in the state $|\psi_{\text{initial}}\rangle = \frac{1}{\sqrt{2}} (a_D^\dagger + a_U^\dagger)|\text{vac}\rangle$. In doing so, we bring the gradient close to zero but eventually set it to a small finite value for the remainder of the experiment. This empirically chosen value compensates for any other sources of tilt in the double-well system and maintains the balanced population in the $|U\rangle$ and $|D\rangle$ states until the experiment is started.

### 3.2 Quench Dynamics with a Gauge Field

After the preparation of the initial state (either one or two particles), the dynamics are initiated by a quench in both directions. In the two-particle case, a double-well potential whose minima align with those of the $y$-lattice is superimposed on the lattice to create a $2 \times N$ confining potential. Afterwards, complex tunneling along the $x$-direction is engineered by ramping on a physical magnetic field gradient while still maintaining a deep optical lattice. This gradient is large enough to suppress tunneling even after the next step, during which the optical lattice is ramped down to a lower depth. It is noteworthy that the resonance frequency for restoring the tunneling is calibrated at this lower lattice depth, since optical potentials of different strengths can have residual gradients resulting in a lattice-depth dependent resonance condition. To restore the tunneling suppressed by the tilt, the Raman lattice power is first ramped up to $V_R \approx 0.05E_r$, then the optical lattice along the $x$-direction is ramped down to $\approx 4E_r$ to increase the bare tunneling in this direction. Afterwards, the frequency of the Raman lattice is chirped to the resonant frequency to enable complex tunneling and thereby realize the Harper-Hofstadter Hamiltonian on the $2 \times N$ ladder that is defined by the bare lattice and the remaining DMD potential. In the two-particle case, the $y$-lattice is additionally quenched to $11E_r$ at the time the Raman lattice is chirped to resonance. In the single-particle case, this is unnecessary since the lattice is left at $11E_r$ from the Landau-Zener sweep. After a given quench time all lattices are ramped to their maximum depth to suppress all dynamics and the atoms are then imaged with single-site resolution (S9). For all two-particle experiments conducted, the doublon fraction at any time during the evolution is sufficiently small such that the results are not affected by the loss of atoms due to parity projection.

### 4 Single-particle experiments

#### 4.1 Initial state fidelity

The initial state for the single-particle experiments would ideally correspond to $|\psi_{\text{initial}}\rangle = \frac{1}{\sqrt{2}} (|U\rangle + |D\rangle)$. However, errors in the adiabaticity of the Landau-Zener ramp can result in either a population imbalance between the two wells or an additional relative phase. Generally then, this state can be written as $|\psi_{\text{initial}}\rangle = \sin(\theta) |U\rangle + \cos(\theta) e^{i\phi} |D\rangle$ where the desired state has $\theta/\pi = 0.25, \phi/\pi = 0$. To measure the population and phase imbalance of the initial state, we let the initial state evolve under just the rung dynamics at a lattice depth of $(V_x, V_y) \approx (4.6)E_r$. Despite the low lattice depth in the $x$ direction, tunneling in this direction is suppressed by the large tilt. The Raman beams that are used in the experiment to restore this tunneling are left off for this measurement. If both the amplitude and the phase were correctly balanced, then the evolution would show no dynamics between the sides of the initial rung and would have a single-well occupancy of 50%. The fitted evolution is shown in Fig. S11 and yields $\theta/\pi = 0.24(1), \phi/\pi = -0.02(2)$. These values are almost equal to the ideal values mentioned above and provide a good calibration of the initial state preparation for the single-particle experiments where an additional phase shift in $\phi$ can arise from the rapid turn-on of the Raman beams.

#### 4.2 Dynamics along the $x$ direction

The chiral nature of the single-particle dynamics is evident from the particle’s center-of-mass motion along the rung direction, plotted separately for each ladder half in figure 3 of this paper. Figure S12 shows the complementary plot for the $x$ direction, describing the expansion of the single-particle wave function along the ladder. The data shows excellent agreement with theory for both ladder halves, implying a low amount of disorder in the system.
Figure S10: **Initial state preparation**: The steps for preparing the one- or two-particle states are shown in part a and b for the x- and y-directions, respectively. i,ii illustrate the superposition of the confining DMD potential and the bare lattice potential. iii,iv illustrate the removal of unwanted atoms from the system due to the removal of the optical lattice and the addition of an anti-confining potential. v,vi illustrate the reloading of the optical lattice and the removal of the additional “cutting” potential from the DMD. The preparation of the delocalized single-particle initial state is illustrated in panel c. The left plot shows the energy of the two instantaneous eigenstates of the system as a function of $\Delta/J$ where the color denotes the overlap of the bare states $|U\rangle$ and $|D\rangle$ with these eigenstates. The initial state for the single-particle experiments is obtained by starting at $\Delta/J \approx 20$ and adiabatically following the ground state to the $\Delta = 0$ point in the diagram, as denoted by the dashed black line in panel b. The sketches above the plot visualize the approximate description of the system at given ratios of $\Delta/J$. The overall preparation sequence is visualized in the additional sketches i-iv. i,ii show the tilting of the lattice while tunneling is still strongly suppressed. iii shows the restoration of tunneling after the depth of the tilted lattice has been reduced. iv shows the final state produced by the Landau-Zener sweep after the tilt has been ramped down. The overall sequence thus corresponds to the state transformation $|U\rangle \rightarrow \frac{1}{\sqrt{2}}(|U\rangle + |D\rangle)$. 
Figure S11: **Preparation of delocalized single-particle states**: Probability to find the particle on the upper well over time. The curve describes the evolution of the fitted initial state $|\psi_{\text{initial}}\rangle = \sin(\theta)|U\rangle + \cos(\theta)e^{i\phi}|D\rangle$ with $\theta/\pi = 0.24(1)$ and $\phi/\pi = -0.02(2)$.

Figure S12: **Single-particle dynamics along the x direction**: We separately determine the center-of-mass coordinates of the single-particle wavefunction for each half of the ladder (See Figure 3). The plot shows the x-coordinate of this center-of-mass versus time.

4.3 Fitting the dynamics

The solid lines shown in Figure 3b are derived from an exact diagonalization of the Harper-Hofstadter with tunneling parameters $J$ and $K$ at flux $\Phi$ for a quench from an initial state, $|\psi_{\text{initial}}\rangle = \sin(\theta)|U\rangle + \cos(\theta)e^{i\phi}|D\rangle$.

The parameters $J$, $K$, $\theta$, and $\phi$ are free parameters while the flux per plaquette $\Phi$ is obtained from an independent calibration. Using exact diagonalization, we can obtain the density distribution at the experimentally used evolution times. The root-mean-square difference between the simulated and measured density distributions is minimized by varying the free parameters. At the optimal parameter values, we compute the center-of-mass of the simulated density distributions, which are plotted in Figure 3b.

5 Heating rates in a Raman lattice

We estimate heating rates in our Raman lattice by measuring the atom number decay as a function of time for different fluxes $\Phi$. Our experiments are performed on a time scale where this exponential decay is entirely dominated by its linear term, yielding

$$N(t) = N_0 \exp\left(-\frac{t}{t_0}\right) \approx N_0 - \frac{N_0}{t_0} t,$$
where \( N_0 \) and \( t_0 \) are the initial atom number and the \( 1/e \) decay time, respectively. Using these quantities as free parameters, we fit individual decay curves to our experimental data, the results of which are shown in Fig. S13.

Due to a variation of single-particle fidelities in the corresponding Mott insulators, the initial (average) atom number varies for different flux values \( \Phi \), but the subsequent decay of the population is given by the heating dynamics in the Raman lattice. For the single-particle case with \( \Phi = \pi/3 \), the fitted decay time scale cannot be distinguished from our vacuum-limited lifetime within the statistical error. For the experiments with two interacting particles in the ladder, we can compare the case without Raman lattice (\( \Phi = 0 \)) to the case with photon-assisted tunneling for flux values \( \Phi = \pi/3, -\pi/3, \pi/2 \) and \( \pi \) (cf. Fig. S14).

Figure S13: **Individual decay curves**: The figure shows the average atom number plotted against time for six experimental sequences realizing different flux values \( \Phi \). For each sequence, a linear function is fitted to the data and the extracted slope is interpreted as the first order expansion of the exponential atom number decay in the system.
Figure S13: Individual decay curves: The figure shows the average atom number plotted against time for six experimental sequences realizing different flux values $\Phi$. For each sequence, a linear function is fitted to the data and the extracted slope is interpreted as the first order expansion of the exponential atom number decay in the system.

Figure S14: Atom number decay rates: The fitted decay rates for the five two-particle experiments plotted as a function of flux. Solid line is the average of decay rate for those cases where flux is non zero.

References


