resonant optical push-out pulse[39,40]. For this, the lattice depths are increased to $V_{s,x}=70(2)E_{r,s}$, $V_{l,x}=30(1)E_{r,l}$, $V_{l,y}=70(2)E_{r,l}$ and $V_z=100(3)E_{r,z}$ over 5 ms to maximize the on-site interaction energy. The atoms, which were initially in the $(F=1, m_F=-1)$ hyperfine state, were converted to $(F=1, m_F=0)$ by using an adiabatic radio-frequency transfer. Here, F denotes the total angular momentum of the atoms. By ramping a magnetic offset field in the presence of a microwave field, we performed a Landau–Zener sweep that adiabatically converted pairs of $m_F=0$ atoms on the same lattice site to an $m_F=+1$ and an $m_F=-1$ atom via coherent spin-changing collisions. The $m_F=-1$ atoms were subsequently removed via an adiabatic microwave transfer to $(F=2, m_F=-2)$, which was followed by a resonant optical pulse after lowering the lattices to $V_{s,x}=0E_{r,s}$, $V_{l,x}=30(1)E_{r,l}$, $V_{l,y}=40(1)E_{r,l}$ and $V_z=40(1)E_{r,z}$.

Sequence for pumping. The superlattice phase can be controlled by slightly changing the frequency of the lasers used for generating the long lattices and thereby moving the relative position between the short and long lattices at the position of the atoms. The pumping along x is performed by slowly changing φ_x, starting from the staggered configuration at $\varphi_x=0.000(5)\pi$, in which the energy difference between neighbouring sites ($|\Delta_x|$) is largest and the tunnel couplings are equal ($\delta J_x=0$). To minimize non-adiabatic transitions to higher bands, each pump cycle consists of three S-shaped ramps: $\varphi_x\in[0, 0.5\pi]$, $[0.5\pi, 1.5\pi]$ and $[1.5\pi, 2\pi]$. This reduces the ramp speed in the vicinity of the symmetric double-well configuration ($\Delta_x=0$) at $\varphi_x=(l+1/2)\pi$, with $l\in\mathbb{Z}$, at which the gap to the first excited band is smallest. The duration of the $\pi/2$ ramps is 7 ms, and 14 ms for the ramp by π. Owing to the limited tuning range of a single laser, a second laser is required for implementing multiple pump cycles, which is set to a constant phase of $\varphi_x=0.000(5)\pi$. At the end of each cycle, an instantaneous switch from the primary laser to the secondary one is made, and within 5 ms the frequency of the former is ramped back to its initial value, corresponding to an identical lattice configuration. After switching back to the first laser, the next cycle continues as described above. We checked experimentally that this handover between the two lasers does not create any measurable band excitations.

Measuring the *in situ* position. To determine the nonlinear COM displacement along y, a double-differential measurement was conducted to minimize the effect of shot-to-shot fluctuations of the atom position. To do this, the COM position is measured before (y_i) and after (y_f) the pumping and compared to a reference sequence ($y_i^{(0)}$, $y_f^{(0)}$). For the latter measurement, the pumping is performed with only the short lattice along y (at $V_{s,y}=40(1)E_{r,s}$); there is therefore no nonlinear response. The initial position is obtained during the doublon removal sequence, where the atoms are initially prepared in the $(F=1, m_F=0)$ hyperfine state and one atom from each doubly occupied site is transferred to $(F=2, m_F=-2)$ using microwave-dressed spin-changing collisions (see above). In addition, we transfer 50% of the atoms on singly occupied sites to the $F=2$ manifold, by applying a microwave π pulse resonant on the $(F=1, m_F=0)\rightarrow(F=2, m_F=0)$ transition. The $F=2$ atoms thus have the same density distribution as the remaining $F=1$ atoms and are imaged before the push-out pulse, which removes them from the lattice. The motion of the atoms due to the nonlinear response is then $\Delta y=(y_f-y_i)-(y_f^{(0)}-y_i^{(0)})$. The difference in the COM displacement along y between θ_1 and θ_2 is defined as $\Delta r_y=\Delta y(\theta_1)-\Delta y(\theta_2)$. For the x direction, it is obtained from $\Delta x=(x_f-x_i)-\delta\bar{x}$ directly without comparing it to the reference sequence. Here, $\delta\bar{x}$ is the average displacement of all data for a given angle, accounting for a small constant offset between the measured initial and final positions.

Relation between COM position and double-well imbalance. If there are no inter-double-well transitions along y, then the change in the double-well imbalance $\delta\mathcal{I}_y=\mathcal{I}_y(\varphi_x)-\mathcal{I}_y(\varphi_x=0)$ can be related directly to the COM motion along y. The COM position in the y direction is

$$y_{\mathrm{COM}}=\frac{d_l}{N}\sum_{ij}\left[(j-1/4)N_{e,ij}+(j+1/4)N_{o,ij}\right]$$

where the sum is over all unit cells, $N_{e,ij}$ ($N_{o,ij}$) is the occupation of the even (odd) sites along y in the (i,j)th unit cell and N is the total number of atoms. Expressing this in terms of the total number of atoms on even and odd sites, $N_e=\sum_{ij}N_{e,ij}$ and $N_o=\sum_{ij}N_{o,ij}$, and assuming that there are no transitions between neighbouring unit cells along y (that is, $\sum_i(N_{e,ij}+N_{o,ij})$ remains constant), the change in the COM position can be written as $\delta y_{\mathrm{COM}}=y_{\mathrm{COM}}(\varphi_x)-y_{\mathrm{COM}}(\varphi_x=0)=d_l\delta\mathcal{I}_y/4$. Note that this derivation implicitly assumes that the COM of the maximally localized Wannier functions on the lattice sites along y is independent of φ_y, which is a valid approximation deep in the tight-binding regime; otherwise, the proportionality factor $d_l/2$ has to be replaced by the distance between the COM of the Wannier functions on the even and odd sites of a double well.

Direct determination of the second Chern number. To determine the second Chern number directly from the measured double-well imbalance $\mathcal{I}_y(\varphi_x)$, the average change in the imbalance per cycle for the entire cloud $\delta\mathcal{I}_y(\varphi_y^{(0)})$ is obtained from a linear fit of the differential imbalance $\mathcal{I}_y(\varphi_x)-\mathcal{I}_y(-\varphi_x)$ for each value of $\varphi_y^{(0)}$. The influence of the excitations can be reduced by restricting the fitting region to a small number of pump cycles. The response of an infinite system is reconstructed by averaging $\delta\mathcal{I}_y(\varphi_y^{(0)})$ over $\varphi_y^{(0)}$ using linear interpolation between the data points. When taking into account all points with $\varphi_x/(2\pi)\leq 3$, this gives $\nu_2^{\mathrm{exp}}=0.84(17)$ for the data in Fig. 3. Note that the linear interpolation for the discrete sampling used in Fig. 3c leads to a systematic shift in ν_2^{exp} of $+0.05$. When correcting for the finite pumping efficiency along x (see below), which can be measured independently without prior knowledge about the system, we obtain $\nu_2^{\mathrm{exp}}=0.94(19)$.

Model for double-well imbalance including experimental imperfections. To isolate the nonlinear response of the lowest band from the band-mapping data, we use a simple model that takes into account band excitations and double occupation of plaquettes, and the experimental pumping efficiency of the linear response. The average double-well imbalance $\mathcal{I}_y(\varphi_x)$ can be written as

$$\mathcal{I}_y(\varphi_x)=n_{\mathrm{gs}}\mathcal{I}_y^{\mathrm{gs}}(\varphi_y)+n_{\mathrm{exc}}\mathcal{I}_y^{\mathrm{exc}}(\varphi_y)+n_2\mathcal{I}_y^{2,\mathrm{gs}}(\varphi_y)$$

where n_{gs} (n_{exc}) is the fraction of atoms on singly occupied plaquettes in the ground (first excited) state along y and n_2 is the fraction of atoms on doubly occupied plaquettes, which we assume to be in the ground state. These quantities can be determined experimentally at each point in the pumping sequence. $\mathcal{I}_y^{\mathrm{gs}}$, $\mathcal{I}_y^{\mathrm{exc}}$ and $\mathcal{I}_y^{2,\mathrm{gs}}$ denote the imbalances of the corresponding states, which depend on the local phase of the y superlattice at the position of the cloud along x, $\varphi_y(x_{\mathrm{COM}})$. The imbalance curves can be calculated theoretically using the respective double-well Hamiltonian (equations (5) or (6)) and can be obtained experimentally by studying the linear pumping response. The COM position in turn depends on the pump parameter φ_x and includes corrections for the finite pumping efficiency

$$x_{\mathrm{COM}}(\varphi_x)=\mathrm{sgn}(\varphi_x)\sum_{i=1}^{|\varphi_x|/\pi}(2\beta_0\beta^i-\beta)d_s$$

for $\varphi_x/\pi\in\mathbb{Z}$. Here, $\beta_0=0.980(4)$ is the initial ground-state occupation along x and $\beta=0.986(2)$ is the pumping efficiency, given by the fraction of atoms that remain in the lowest subband during each half of a pump cycle and are therefore transferred by one lattice site along x. The main contributions that limit the pumping efficiency are band excitations in the pumping direction and non-adiabatic transitions between neighbouring double wells induced by the external harmonic confinement. Although the local slope of the transverse response for doubly occupied plaquettes differs from that for single atoms, they exhibit the same quantized transport along x and y for the parameters used in the experiment when covering the entire 4D pump path.

Measuring band excitations. Band excitations in the y direction are measured by adiabatically ramping the superlattice phase $\varphi_y^{(0)}$ from its initial value to $\pi/2\pm0.156(5)\pi$ and subsequently increasing the short lattice depth to $V_{s,y}=40(1)E_{r,s}$. In this lattice configuration, ground-state atoms on singly and doubly occupied plaquettes are fully localized on the lower-lying site along y, owing to the large double-well tilt Δ_y and the suppression of tunnelling as $J_y, \delta J_y\rightarrow 0$. On the other hand, atoms in the excited band along y localize on the higher-lying site and can be detected directly by measuring the resulting double-well imbalance.

Detecting doubly occupied plaquettes. The doublon fraction can be determined by taking advantage of the fact that two atoms in the same double well localize on the lower-lying site only at much larger double-well tilts than for a single atom, owing to the repulsive on-site interaction. For this, the double wells along y are first merged into a single site by removing the short lattice and increasing the long lattice to $V_{l,y}=30(1)E_{r,l}$ within 5 ms. At the same time, the orthogonal lattice depths are ramped up to $V_{s,x}=70(2)E_{r,s}$ and $V_z=100(3)E_{r,z}$ to increase the interaction energy. After that, $\varphi_y^{(0)}$ is shifted adiabatically to either $0.474(5)\pi$ or $0.431(5)\pi$ and the sites are again split into double wells by ramping up the short lattice to $V_{s,y}=40(1)E_{r,s}$. At $\varphi_y^{(0)}=0.431\pi$, single atoms and doublons are both fully localized on the lower-lying site. On the other hand, at $\varphi_y^{(0)}=0.474\pi$ single atoms are still very well localized on the lower site, but two atoms in the same double well localize on different sites owing to the large interaction energy $U>\Delta_y$. By determining the site occupations for both phases, we can therefore infer the doublon fraction from the difference in the even–odd imbalance between the two measurements.

Calculating the double-well imbalance along *y*. The measurement of the population imbalance in the y direction as a function of φ_x for Figs 3 and 4 is performed after an integer or half-integer number of pump cycles ($\varphi_x=l\pi$, $l\in\mathbb{Z}$). At these points, the superlattice along x is in the staggered configuration, with the maximum energy offset $|\Delta_x|\gg J_x$ and $\delta J_x=0$. The atoms are thus fully localized on either even