or odd sites along x for $\varphi_x = 2l\pi$ or $\varphi_x = (2l+1)\pi$, respectively. The four-site unit cell of the 2D superlattice therefore effectively reduces to a double well along y.

For singly occupied double wells, the expected imbalance in the y direction for atoms in the ground ($\mathcal{I}_y^{\mathrm{gs}}$) and first excited state ($\mathcal{I}_y^{\mathrm{exc}}$) can then be calculated from the single-particle double-well Hamiltonian

$$\hat{H}_{\mathrm{DW}}^{(1)}(\varphi_y) = \begin{pmatrix} \Delta_y(\varphi_y)/2 & -J_y^0(\varphi_y) \\ -J_y^0(\varphi_y) & -\Delta_y(\varphi_y)/2 \end{pmatrix} \quad (5)$$

with $J_y^0(\varphi_y) = J_y(\varphi_y) + \delta J_y(\varphi_y)/2$ and using the Fock basis for the atoms on even and odd sites, $|1, 0\rangle$ and $|0, 1\rangle$, respectively.

Correspondingly, the imbalance for the ground state of a doubly occupied double well ($\mathcal{I}_y^{2,\mathrm{gs}}$) can be determined using the two-particle double-well Hamiltonian

$$\hat{H}_{\mathrm{DW}}^{(2)}(\varphi_y) = \begin{pmatrix} U + \Delta_y & -\sqrt{2}J_y^0 & 0 \\ -\sqrt{2}J_y^0 & 0 & -\sqrt{2}J_y^0 \\ 0 & -\sqrt{2}J_y^0 & U - \Delta_y \end{pmatrix} \quad (6)$$

in the Fock basis $\{|2, 0\rangle, |1, 1\rangle, |0, 2\rangle\}$. Here, U denotes the on-site interaction energy for two atoms localized on the same lattice site.

Fit function for nonlinear response. On the basis of the above model, the experimental data are fitted with the function $\mathcal{I}_y(\varphi_x) + \mathcal{I}_0$ with $\varphi_y \to \varphi_y^{\mathrm{exp}} = \varphi_y^{(0)} + \alpha(\varphi_y - \varphi_y^{(0)})$. The two fit parameters are the pre-factor α, which describes the change in the superlattice phase along y with φ_x compared to the ideal case $\varphi_y^{\mathrm{exp}} = \varphi_y$, and an overall offset $\mathcal{I}_0$. The transport properties of the lowest band are encoded in the slope of the ground-state imbalance at $\varphi_x = 0$. Knowing α, it can be related to the ideal slope via

$$\frac{\partial \mathcal{I}_y^{\mathrm{gs}}(\varphi_y^{\mathrm{exp}})}{\partial \varphi_x} = \frac{\partial \mathcal{I}_y^{\mathrm{gs}}(\varphi_y^{\mathrm{exp}})}{\partial \varphi_y^{\mathrm{exp}}} \frac{\partial \varphi_y^{\mathrm{exp}}}{\partial \varphi_x} = \alpha \frac{\partial \mathcal{I}_y^{\mathrm{gs}}(\varphi_y)}{\partial \varphi_x}$$

Per cycle, this gives a change in the population imbalance for ground-state atoms of

$$\delta \mathcal{I}_y^{\mathrm{gs}} = \alpha \left[\mathcal{I}_y^{\mathrm{gs}}(\varphi_y)\Big|_{\varphi_x = 2\pi} - \mathcal{I}_y^{\mathrm{gs}}(\varphi_y)\Big|_{\varphi_x = 0} \right]$$

Determining the second Chern number from the scaling of the nonlinear response with $\boldsymbol{\theta}$. The COM displacement per cycle along y for an infinite system, $\delta y_{\mathrm{COM}} = \nu_2 \theta a_x a_y / d_{\mathrm{l},y}$, scales linearly with the perturbing angle θ. The second Chern number can thus be extracted from the slope of $\delta y_{\mathrm{COM}}(\theta)$. Having confirmed that the measured shape of $\delta \mathcal{I}_y^{\mathrm{gs}}(\varphi_y^{(0)})$ is the same as expected theoretically, the response of an infinite system at a given angle θ can be inferred from a single measurement of $\delta \mathcal{I}_y^{\mathrm{gs}}$ at a fixed $\varphi_y^{(0)}$. This holds for all angles because the shape of $\overline{\Omega}(\varphi_y^{(0)})$ is independent of θ. To obtain ν_2, it is therefore sufficient to determine the slope of $\delta \mathcal{I}_y^{\mathrm{gs}}(\theta)$ at a constant $\varphi_y^{(0)}$.

Nonlinear response versus lattice depth. The technique for detecting the nonlinear response with site-resolved band mapping, introduced in the main text, allows us to determine the slope over a wide range of lattice parameters accurately. To demonstrate this, we measure the slope of the nonlinear response at $\varphi_y^{(0)} = 0.500(5)\pi$ and $\theta = 0.54(3)$ mrad for various values of the transverse short-lattice depth $V_{\mathrm{s},y}$ (Extended Data Fig. 1). As expected, the slope increases with larger depths as the band gap decreases and the Berry curvature Ω^y becomes more and more localized around $\varphi_y^{(0)} = (l + 1/2)\pi$ with $l \in \mathbb{Z}$.

At $V_{\mathrm{s},y} = 6.25 E_{\mathrm{r,s}}$, the first and second excited subbands along y touch for $\varphi_y^{(0)} = l\pi$, leading to a topological transition where the signs of the first and second Chern number of the first excited subband change from $+1$ for $V_{\mathrm{s},y} < 6.25 E_{\mathrm{r,s}}$ to -1 for $V_{\mathrm{s},y} > 6.25 E_{\mathrm{r,s}}$. This corresponds to a transition between the Landau and Hofstadter regimes[25]. For the lowest band, the two regimes are topologically equivalent and the atoms therefore move in the same direction. In both limits, the experimentally determined slope matches very well with the one expected in an ideal system. This nicely illustrates that the transport properties of the lowest band can be extracted correctly in both regimes, even in the presence of atoms in the first excited band.

Alignment of the tilted superlattice. Each optical lattice is created by retroreflecting a laser beam, which is focused onto the atoms by a lens on either side of the cloud. For the superlattices, the incoming beams of the short and long lattices are overlapped using a dichroic mirror in front of the first lens. To control the tilt angle θ of the long lattice along y, a glass block is placed in the beam path before the overlapping. By rotating this glass block, a parallel displacement of the incoming beam can be induced, which is then converted into an angle θ relative to the short lattice beam at the first lens. The two beams intersect at the focus point of the lens, which corresponds to the position of the cloud of atoms. After passing through the second lens behind the cloud, both beams are retroreflected by the same mirror. The counter-propagating beams travel along the paths of the incoming beams, thereby creating the lattice potentials with the same relative angle θ.

Determining the angle $\boldsymbol{\theta}$. When the long lattice in the y direction is tilted by an angle θ with respect to the short lattice, the phase of the superlattice along y depends on the position along x. This leads to a modification of the on-site potential, which for small angles can be approximated as a linear gradient along the x axis, pointing in opposite directions on even and odd sites in y

$$\Delta_y^{m_y}(\varphi_y) \approx \Delta_y^{m_y}(\varphi_y^{(0)}) + (-1)^{m_y} \delta m_x$$

The strength of the gradient is

$$\delta = \frac{\pi d_{\mathrm{s}}}{d_{\mathrm{l}}} \frac{\partial \Delta_y}{\partial \varphi_y}\bigg|_{x=0} \theta$$

for a given superlattice phase $\varphi_y^{(0)}$ and can therefore be used to determine θ. To do this in the experiment, a superfluid is prepared at $\boldsymbol{k} = \mathbf{0}$ in a 2D lattice with $V_{\mathrm{s},x} = 13.0(4) E_{\mathrm{r,s}}$ and $V_{\mathrm{l},y} = 10.0(3) E_{\mathrm{r,l}}$. After increasing $V_{\mathrm{l},y}$ to $70(2) E_{\mathrm{r,l}}$ within 0.2 ms, the lattice sites are split along y by ramping up the short lattice in the y direction to $V_{\mathrm{s},y} = 20.0(6) E_{\mathrm{r,s}}$ in 0.4 ms. The superlattice phase $\varphi_y^{(0)}$ is set to either $0.344(5)\pi$ or $0.656(5)\pi$ such that the atoms fully localize on even or odd sites along y, respectively. The resulting Bloch oscillations that are induced by the gradient are probed by measuring the momentum distribution of the atoms after a variable hold time. The angle θ is then calculated from the average Bloch oscillation period of both phases to minimize the influence of additional residual gradients.

Data availability. The data that support the findings of this study are available from the corresponding author on reasonable request.

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