

METHODS

Hall response of the 4D quantum Hall system. Assuming perfect adiabaticity, the Hall response of the 4D system shown in Fig. 1a, b can be evaluated from the semi-classical equations of motion for a wave packet centred at position \mathbf{r} and quasi-momentum \mathbf{k} (ref. 32)

$$\dot{\mathbf{r}}^\mu = \frac{1}{\hbar} \frac{\partial \mathcal{E}(\mathbf{k})}{\partial k_\mu} + \dot{\mathbf{k}}_\nu \Omega^{\nu\mu}(\mathbf{k})$$

$$\hbar \dot{\mathbf{k}}_\mu = qE_\mu + q\dot{\mathbf{r}}^\nu B_{\mu\nu}$$

Here, $\mathcal{E}(\mathbf{k})$ is the energy of the respective eigenstate at \mathbf{k} , q is the charge of the particle and Einstein notation is used for the spatial indices $\mu, \nu \in \{w, x, y, z\}$. The orientation of the axes in Fig. 1a, b is chosen such that the 4D Levi-Civita symbol is $\varepsilon_{wxyz} = +1$. The velocity of the wave packet $\mathbf{v} = \dot{\mathbf{r}}$ has two contributions: the group velocity, which arises from the dispersion of the band, and the anomalous velocity, which is due to the non-zero Berry curvature

$$\Omega^{\nu\mu}(\mathbf{k}) = i(\langle \partial_{k_\nu} u | \partial_{k_\mu} u \rangle - \langle \partial_{k_\mu} u | \partial_{k_\nu} u \rangle)$$

For a filled or homogeneously populated band, the group velocity term vanishes and with $\mathbf{E} = E_z \mathbf{e}_z$ and $\mathbf{B} = \mathbf{0}$ the linear Hall response is given by the COM velocity

$$\mathbf{v}_{\text{COM}}^{(0)} = \frac{q}{h} A_M^{\text{zx}} E_z \nu_1^{\text{zx}} \mathbf{e}_x$$

where A_M^{zx} denotes the size of the magnetic unit cell in the x - z plane and

$$\nu_1^{\text{zx}} = \frac{1}{2\pi} \oint_{\text{BZ}} \Omega^{\text{zx}} d^2k$$

denotes the first Chern number of the 2D quantum Hall system in the x - z plane. The integration is performed over the 2D Brillouin zone (BZ) spanned by k_x and k_z .

Adding the perturbing magnetic field B_{zw} generates a Lorentz force that acts on the moving cloud²⁰, $\hbar \dot{\mathbf{k}} = qE_z \mathbf{e}_z - qv_x^{(0)} B_{\text{zw}} \mathbf{e}_w$. (This additional force can alternatively be interpreted as arising from a Hall voltage in the w direction that is created by the current along x in the presence of B_{zw} .) This force in turn induces an additional anomalous velocity along y , giving rise to the nonlinear Hall response. The resulting average velocity is then

$$\mathbf{v}_{\text{COM}} = \frac{q}{h} A_M^{\text{zx}} E_z \nu_1^{\text{zx}} \mathbf{e}_x - \left(\frac{q}{h} \right)^2 A_M E_z B_{\text{zw}} \nu_2^{\text{zy}} \mathbf{e}_y$$

with A_M the size of the 4D magnetic unit cell. The second Chern number is given by

$$\nu_2 = \frac{1}{4\pi^2} \oint_{\text{BZ}} \Omega^{\text{xw}} \Omega^{\text{zy}} + \Omega^{\text{xy}} \Omega^{\text{wz}} + \Omega^{\text{zx}} \Omega^{\text{wy}} d^4k$$

where BZ denotes the 4D Brillouin zone.

Tight-binding Hamiltonian of the 2D superlattice. In the tight-binding limit, the motion of non-interacting atoms in a 2D superlattice is captured by the following Hamiltonian

$$\begin{aligned} \hat{H}_{2\text{D}}(\varphi_x, \varphi_y) = & \\ & - \sum_{m_x, m_y} [J_x(\varphi_x) + \delta J_x^{m_x}(\varphi_x)] \hat{a}_{m_x+1, m_y}^\dagger \hat{a}_{m_x, m_y} + \text{h.c.} \\ & - \sum_{m_x, m_y} [J_y(\varphi_y) + \delta J_y^{m_y}(\varphi_y)] \hat{a}_{m_x, m_y+1}^\dagger \hat{a}_{m_x, m_y} + \text{h.c.} \\ & + \sum_{m_x, m_y} [\Delta_x^{m_x}(\varphi_x) + \Delta_y^{m_y}(\varphi_y)] \hat{a}_{m_x, m_y}^\dagger \hat{a}_{m_x, m_y} \end{aligned} \quad (1)$$

Here, $\hat{a}_{m_x, m_y}^\dagger$ (\hat{a}_{m_x, m_y}) is the creation (annihilation) operator acting on the (m_x, m_y) th site in the x - y plane. The first (second) term describes the hopping between neighbouring sites along the x axis (y axis), with tunnelling matrix elements $J_\mu + \delta J_\mu^{m_\mu}$, with $\mu \in \{x, y\}$. The last term contains the on-site potential of each lattice site, $\Delta_x^{m_x} + \Delta_y^{m_y}$. In the presence of the long lattices, the tunnel couplings and on-site energies are modulated periodically by $\delta J_\mu^{m_\mu}$ and $\Delta_\mu^{m_\mu} + \Delta_y^{m_y}$, respectively. Both modulations depend on the respective superlattice phases φ_μ .

For the lattice configuration used in the experiment, where $d_{1,\mu} = 2d_{s,\mu}$, these modulations can be expressed as $(-1)^{m_\mu} \delta J_\mu/2$ and $(-1)^{m_\mu} \Delta_\mu/2$, and equation (1) reduces to the 2D Rice-Mele Hamiltonian²⁷

$$\begin{aligned} \hat{H}_{2\text{D}}(\varphi_x, \varphi_y) = & \\ & - \sum_{m_x, m_y} [J_x(\varphi_x) + (-1)^{m_x} \delta J_x(\varphi_x)/2] \hat{a}_{m_x+1, m_y}^\dagger \hat{a}_{m_x, m_y} + \text{h.c.} \\ & - \sum_{m_x, m_y} [J_y(\varphi_y) + (-1)^{m_y} \delta J_y(\varphi_y)/2] \hat{a}_{m_x, m_y+1}^\dagger \hat{a}_{m_x, m_y} + \text{h.c.} \\ & + \sum_{m_x, m_y} \frac{1}{2} [(-1)^{m_x} \Delta_x(\varphi_x) + (-1)^{m_y} \Delta_y(\varphi_y)] \hat{a}_{m_x, m_y}^\dagger \hat{a}_{m_x, m_y} \end{aligned}$$

Mapping a 2D topological charge pump to a 4D quantum Hall system. The Hamiltonian of a 2D topological charge pump for a given set of parameters $\{\varphi_x, \varphi_y\}$, $\hat{H}_{2\text{D}}(\varphi_x, \varphi_y)$, can be interpreted as a Fourier component of a higher-dimensional quantum Hall system. Using the approach of dimensional extension^{6,22}, a 2D charge pump can be mapped onto a 4D quantum Hall system, the Fourier components of which are sampled sequentially during a pump cycle. This is demonstrated in the following for the deep tight-binding limit $V_{s,\mu} \gg V_{l,\mu}^2/(4E_{\text{r,s}})$, $\mu \in \{x, y\}$, in which the corresponding 4D system consists of two 2D Harper-Hofstadter-Hatsugai models³³⁻³⁶ in the x - z and y - w planes. A similar analogy can be made in the opposite limit of vanishingly short lattices, $V_{s,x} \rightarrow 0$ and $V_{s,y} \rightarrow 0$. In this case, each axis of the 2D lattice maps onto the Landau levels of a free particle in an external magnetic field in two dimensions²⁵. For the lowest band, these two limiting cases are topologically equivalent; that is, they are connected by a smooth crossover without closing the gap to the first excited band. The topological invariants that govern the linear and nonlinear response are thus independent of the depth of the short lattices.

In the deep tight-binding regime, J_x and J_y become independent of the superlattice phases and the modulations can be approximated as³⁷

$$\begin{aligned} \delta J_x^{m_x}(\varphi_x) &= \frac{\delta J_x^{(0)}}{2} \cos[\tilde{\Phi}_{\text{zx}}(m_x + 1/2) - \varphi_x] \\ \delta J_y^{m_y}(\varphi_y) &= \frac{\delta J_y^{(0)}}{2} \cos[\tilde{\Phi}_{\text{yw}}(m_y + 1/2) - \varphi_y] \\ \Delta_x^{m_x}(\varphi_x) &= -\frac{\Delta_x^{(0)}}{2} \cos(\tilde{\Phi}_{\text{zx}} m_x - \varphi_x) \\ \Delta_y^{m_y}(\varphi_y) &= -\frac{\Delta_y^{(0)}}{2} \cos(\tilde{\Phi}_{\text{yw}} m_y - \varphi_y) \end{aligned}$$

with $\tilde{\Phi}_{\text{zx}} = 2\pi d_{s,x}/d_{l,x}$ and $\tilde{\Phi}_{\text{yw}} = 2\pi d_{s,y}/d_{l,y}$, $\delta J_\mu^{(0)}$ and $\Delta_\mu^{(0)}$ denote the modulation amplitudes, which are determined by the lattice depths. In this case, $\hat{H}_{2\text{D}}$ is equivalent to the generalized 2D Harper model³³, which describes the Fourier components of a 4D lattice model with two uniform magnetic fields in orthogonal subspaces. The 4D parent Hamiltonian is obtained via an inverse Fourier transform⁶

$$\hat{H}_{4\text{D}} = \frac{1}{4\pi^2} \int_0^{2\pi} \hat{H}_{2\text{D}}(\varphi_x, \varphi_y) d\varphi_x d\varphi_y^{(0)}$$

with

$$\begin{aligned} \hat{a}_{m_x, m_y}^\dagger &= \sum_{m_z, m_w} e^{i(\varphi_x m_z + \varphi_y^{(0)} m_w)} \hat{a}_{\mathbf{m}}^\dagger \\ \hat{a}_{m_x, m_y} &= \sum_{m_z, m_w} e^{-i(\varphi_x m_z + \varphi_y^{(0)} m_w)} \hat{a}_{\mathbf{m}} \end{aligned}$$

and where $\mathbf{m} = \{m_x, m_y, m_z, m_w\}$ indicates the position in the 4D lattice. This yields

$$\hat{H}_{4\text{D}} = \hat{H}_{\text{zx}} + \hat{H}_{\text{yw}} + \hat{H}_{\delta J}$$

The first term (\hat{H}_{zx}) describes a 2D Harper-Hofstadter model³³⁻³⁵ in the x - z plane with a uniform magnetic flux per unit cell, $\Phi_{\text{zx}} = \Phi_0 \tilde{\Phi}_{\text{zx}}/(2\pi) = (d_{s,x}/d_{l,x}) \Phi_0$, with Φ_0 denoting the magnetic flux quantum

$$\begin{aligned} \hat{H}_{\text{zx}} = & - \sum_{\mathbf{m}} J_x \hat{a}_{\mathbf{m}+\mathbf{e}_x}^\dagger \hat{a}_{\mathbf{m}} + \text{h.c.} \\ & - \sum_{\mathbf{m}} \frac{\Delta_x^{(0)}}{4} e^{i\tilde{\Phi}_{\text{zx}} m_x} \hat{a}_{\mathbf{m}+\mathbf{e}_x}^\dagger \hat{a}_{\mathbf{m}} + \text{h.c.} \end{aligned}$$

Correspondingly, the second term (\hat{H}_{yw}) is an independent 2D Harper-Hofstadter model in the y - w plane with $\Phi_{\text{yw}} = (d_{s,y}/d_{l,y}) \Phi_0$. Owing to the positional dependence