Multiband topological phases of periodically kicked molecules

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We show that the simplest of existing molecules – closed-shell diatomics not interacting with one another – host topologically nontrivial phases when driven by periodic far-off-resonant laser pulses. A periodically kicked molecular rotor can be mapped onto a “crystalline” lattice in angular momentum space. This allows to define quasi-momenta and the band structure in the Floquet representation, by analogy with the Bloch waves of solid-state physics. Applying laser pulses spaced by 1/3 of the molecular rotational period creates a lattice with three atoms per unit cell with staggered hopping, whose band structure features Dirac cones. These Dirac cones, topologically protected by reflection and time-reversal symmetry, are reminiscent of (although not equivalent to) the ones seen in graphene. They – and the corresponding edge states – are broadly tunable by adjusting the laser intensities and can be observed in present-day experiments by measuring molecular alignment and populations of rotational levels. This paves the way to study controllable topological physics in gas-phase experiments with small molecules as well as to classify dynamical molecular states by their topological invariants.

The quantum nature of electrons in solids gives rise to a number of fascinating phenomena, such as the quantum Hall effect, that are ultimately related to the geometric and topological properties of the Brillouin zone [1]. Topological phases are characterized by their topological invariants and show remarkable properties, e.g., quantized transport or the bulk-boundary correspondence [2]. Since the early works of Thouless [3] and Haldane [4, 5] the field has drastically expanded, leading to discoveries of several topological phases of matter, such as topological insulators, Weyl semimetals, and topological superconductors [6–8].

Unlike the translational motion of an electron in a lattice, rotations of a molecule correspond to the non-Abelian group SO(3). While free rotations basically correspond to trivial paths in that manifold [9, 10], in this Letter we show that laser pulses can guide the molecule along topologically nontrivial paths, allowing for nonzero Berry phases and alike. In particular, we explore the similarities between the Bloch theorem (for a system periodic in space) and the Floquet theorem (for a system periodic in time) to show that a molecule driven by periodic laser pulses can be mapped onto a translationally invariant hopping model hosting nontrivial topology. Although the ideas of topology in molecules have been extensively exploited in the context of conical intersections of potential energy surfaces in real space [11, 12] (including light-induced conical intersections [13, 14]), the results presented here allow to directly bridge the ideas of topologically protected phases in condensed-matter physics with the realm of molecules.

At energies well below electronic and vibrational excitations diatomic molecules essentially behave as rigid linear rotors [15]. A two-dimensional kicked rotor is a paradigmatic model used to study nonlinear dynamics, dynamical localization and quantum chaos [16, 17]. Since the original works of Casati and Chirikov [18–21], the predictions of the theory have been verified in several experiments, e.g., with atoms in microwave fields [22, 23], Rydberg atoms [24], atomic matter waves [25], and Bose-Einstein condensates [26]. Topological aspects of double kicked two-dimensional rotors have also been extensively explored in connection to spectra resembling the Hofstadter butterfly and the corresponding Chern numbers [27–29]. Periodically driven three-dimensional molecular rotors have been studied theoretically with a particular focus on resonances and Anderson localization [30–32] and edge states [33, 34]. Several phenomena, such as quantum resonances [35], Bloch oscillations [36, 37] and dynamical localization [38–40] have already been observed in experiments with molecules. Moreover, recent advances in imaging of molecular rotational dynamics [41–43] and control of their angular degrees of freedom [44, 45] open new possibilities to probe kicked rotor physics. While these advances are particularly important for the understanding of reactions and other fundamental processes in physical chemistry [46, 47], they can – as we show below – find applications in other seemingly unrelated areas of physics, such as the study of topological phases.

In this Letter we demonstrate that apart from the rich physics related to transport and localization, driving even the simplest molecules by specifically designed periodic laser pulses allows to probe the nontrivial topology of their rotational states. Here we engineer an effective topological semimetal with linear dispersing topological edge states, however, more involved models characterized by other topological invariants can potentially be realized as well. In addition to a higher degree of control achievable in experiment, periodically kicked molecules are able to form multiband topological systems. This paves the way to realize non-Abelian topological phases, whose study in solid-state settings has been quite limited so far.
In what follows, we consider the simplest case of a linear closed-shell molecule which is periodically kicked by a far-off-resonant, linearly polarized laser. The general idea, however, is straightforward to extend to more complex molecules (e.g., symmetric and asymmetric tops) and to other kinds of fields, which might further expand the range of realizable Hamiltonians. When the laser pulse duration is significantly shorter than the rotational period of the molecule, we can write the Hamiltonian as follows [37, 40, 48]:

\[ \hat{H}_{\text{mol}}(t) = B \hat{L}^2 - \left[ P_1 \cos^2(\hat{\theta}) + P_2 \cos(\hat{\theta}) \right] \sum_{q=0}^{\infty} \delta(t - qT) \equiv \hat{V}(P_1, P_2) \]  

(1)

Here \( B = \pi \hbar / T_{\text{rot}} \) is the molecular rotational constant with \( T_{\text{rot}} \) the rotational period, and the laser pulses are defined by their intensities, \( P_1, P_2 \), and the time period between them, \( T \). Unlike most models taking into account either the \( \cos^2(\hat{\theta}) \) term ("regular" multi-cycle laser pulses) or the \( \cos(\hat{\theta}) \) term (half-cycle laser pulses [48–50]), here we include both, which allows to create tunable Dirac cones, not observable with either one of the terms [51].

Casati et al. [17, 18] have shown that a periodically driven pendulum, and hence also a molecule, displays two regimes: the dynamical localization regime, i.e., localization on a lattice in time (instead of a lattice in space), and the quantum resonance regime, where the pendulum delocalizes. In this work, in order to achieve a banded system, we focus on the resonant regime [35], i.e., \( T = T_{\text{rot}}/N \).

The time-translation operator of the Hamiltonian defined in Eq. (1) after one rotational period takes the form

\[ \hat{U} = e^{-i\frac{\pi \hat{L}^2}{N}} e^{i\hat{V}(P_1, P_2)}. \]  

(2)

The ideas described below are based on the analogy between "real" solid-state systems, which are periodic in space, and molecules periodically kicked in time. Periodically driven systems can be described in terms of the so-called Floquet states, \( |\psi_n\rangle \), and the corresponding quasienergies, \( \epsilon_n \) [52, 53]. These can be defined through the time-translation operator of Eq. (2) as \( \hat{U}|\psi_n\rangle = e^{i\epsilon_n} |\psi_n\rangle \), or, equivalently, through the effective Hamiltonian \( \hat{H} = i \log \hat{U} \) with \( \hat{H}|\psi_n\rangle = \epsilon_n |\psi_n\rangle \). At a quantum resonance, the quasienergies form \( N \) bands, which is a result of the \( N \)-periodicity of the \( e^{-i\frac{\pi \hat{L}^2}{N}} \) operator [54].

It is important to note that characterizing periodically kicked molecules in terms of their Floquet states and quasienergies has been previously done in a number of works, see e.g., Refs. [33, 37]. In what follows, we go one step further and demonstrate that at a quantum resonance one can introduce quasimomenta of periodically driven molecular states. This makes it possible to work with “molecular Bloch bands” and to study their topology, which provides a direct bridge to the condensed matter systems.

In particular, we make use of the fact that for nonzero values of angular momentum, the “hopping” matrix elements between different angular momentum “lattice sites” converges to an approximately constant value, \( H_{\ell\ell'} \approx H_{\ell+N,\ell'+N} \) for \( \ell, \ell' \gg 0 \), with \( H_{\ell\ell'} = \langle \ell | \hat{H} | \ell' \rangle \), cf. Eq. (1) [51]. This allows to create an effective translational invariant tight-binding model where the hopping is controlled by the periodic laser pulses, see Fig. 1. Note that while translational invariance is exact for a planar rotor, it is only approximately achieved for 3D molecular rotors in the limit of \( \ell, \ell' \gg 0 \), which, however, suffices for the purposes of our proposal. Translational invariance within a unit-cell of dimension \( N \) allows us to define the Fourier transform with \( k \in [0, 2\pi) \)

\[ H_{ij}(k) = \sum_{\Delta n} e^{-i\Delta n \cdot k} H_{ij}(\Delta n) \]  

(3)

with \( H_{ij}(\Delta n) = H_{N-n_0+i,N-(n_0+\Delta n)+j} \) and \( n_0 > 0 \), which is important when working with a finite lattice. For an infinite and perfectly translationally invariant system, we would sum over all \( \Delta n \in \mathbb{Z} \). However, here we need to choose \( -n_0 < \Delta n < n_0 \) with a unit-cell \( n_0 \) far away from the boundary in order to exclude edge effects. Note that this finite-size Fourier transform is only approximate and does not capture the physics at the boundary. However, as it turns out, the spectrum obtained within this approximation agrees very well with the results of exact diagonalization and captures the behavior at a quantum resonance, i.e., the indefinite growth of rotational energy. Similarly, for any function in that space we define

\[ f_i(k) = \sum_{\Delta n} e^{-i\Delta n \cdot k} f(i + N(n_0 + \Delta n)). \]  

(4)

Correspondingly, \( H(k)|\psi_n(k)\rangle = \epsilon_n(k)|\psi_n(k)\rangle \) where \( k \) plays the role of quasimomentum of the angular momentum lattice and \( |\psi_n(k)\rangle \in \mathbb{C}^N \) (note that \( H(k) \) and other operators are \( N \times N \) matrices in \( k \)-space).
FIG. 2. Results of exact diagonalization for $N = 3$ and $P = 2.5$, see Eq. (5). (a) The full spectrum with the bulk states shown in orange and the edge states in red. Two linearly dispersing edge states (one for each edge) connect the Dirac cones. (b) The quasienergies $\varepsilon_k$ near the Dirac cone, shown as a function of $k$ and $\alpha$. As a result of time-reversal and reflection symmetries, the two bands touch at $k = \pi$. The Dirac cone is topologically protected and cannot be gapped out by any deformation which preserves the symmetry. (c) The absolute value of the wavefunction of the edge state. (d) A generic bulk state.

Since there are $N$ bands, the Hamiltonian can be written as $H(k) = a(k)I_N + \sum_i d_i(k) \cdot \lambda_i$ in terms of the generalized Gell-Mann matrices $\lambda_i \in \mathrm{SU}(N)$, which form a linearly independent basis of traceless $N \times N$ matrices, and $a(k) \in \mathbb{R}$, $d_i(k) \in \mathbb{R}^{N^2-1}$, see Ref. [55]. The mapping described above connects kicked molecules to single-particle models of condensed matter physics. Furthermore, the symmetries of the effective Hamiltonian are determined by the symmetries of the time-translation operator. Unlike a Hermitian system, however, this system exhibits periodic quasienergies; the free choice of the initial time results in a gauge freedom of the quasienergies. Since the first and the last band can touch over the periodic boundary, this opens up new possibilities such as anomalous topological Floquet insulators [56, 57], which are out of the scope of this Letter.

Here we focus on the case of $N = 3$ for intermediate coupling where no closing of all gaps occurs [33], since this, to the best of our knowledge, is the simplest situation featuring nontrivial topological physics for a kicked molecule. We choose the following parametrization of the laser intensity parameters:

$$ P_1(\alpha) = P \cos^2(\alpha), \quad P_2(\alpha) = P \sin^2(\alpha) \quad (5) $$

in terms of the offset parameter $\alpha \in [0, 2\pi)$, which serves as an effective second dimension in addition to quasi-momentum and laser strength $P$. Since the linearly-polarized laser of Eq. (1) preserves the projection of angular momentum, $m$, we consider only the states with $m = 0$. Fig. 2 shows the results of exact diagonalization. Here we set the cut-off for the real-space diagonalization to $l_{\text{max}} = 500$. There are three bands, shown in Fig. 2(a), with the second and third touching at two points, forming Dirac cones, Fig. 2(b). The Fourier transform shows that the bands touch exactly at $k = \pi$, indicating a symmetry in quasimomentum space. Indeed, the system has a reflection and a time-reversal symmetry that commute with each other [51], i.e.

$$ TH_k = H_{-k}^\ast T \quad \text{and} \quad RH_k = H_{-k}^\ast \mathcal{R}. \quad (6) $$

This implies that for the $k \rightarrow -k$ invariant points, i.e. $k = 0$ or $k = \pi$, the Hamiltonian commutes with $T$ and $\mathcal{R}$. From the 10-fold classification with crystal symmetries the class is $\mathrm{A}_k^\ast \mathcal{R}_+$. The topological analysis of the two Dirac cones proceeds as follows. We combine the two variables into a vector $k = (k, \alpha)$, and compute the Berry connection, $\mathcal{A}_{\alpha}(k) = i\langle \psi_n(k) | \nabla_k \psi_n(k) \rangle$, which is well-defined if we choose a gauge such that the eigenstates form a smooth manifold. Numerically evaluating the Berry phase around one of these cones in the second or third band results in $\int_{\gamma} \mathcal{A}(k)_{\alpha} dk = \pm \pi$ thus proving the topological nature of these cones [59].

As with spinless graphene, we observe two linear dispersing edge states, connecting the two Dirac cones. This is a result of the bulk-boundary correspondence for Dirac semimetals with crystal symmetries. For the bulk states we find plane-wave solutions as predicted by the Fourier-transformed Hamiltonian. The edge states manifest themselves as states localized at the lower end of the lattice ($l \approx 0$). Topologically non-protected edge states in kicked molecules have already been theoretically described [33, 34]. The localized edge states shown in Fig. 2(c) are, on the other hand, topological, in the sense that they cannot be destroyed without merging the two Dirac cones.

The topological characteristics of the band structure shown in Fig. 2 have observable experimental signatures. In order to demonstrate that, we consider the time evolution of a molecule adiabatically driven through the Dirac cones (for fast quenches the phenomenon persists, but is superimposed by the quench dynamics). We adiabatically change the parameter $\alpha$ of Eq. (5) as

$$ \alpha(t) = t \cdot (\alpha_c/t_c) \quad (7) $$
where $\alpha_c$ denotes the critical value of $\alpha$ for which the cones appear at $k_c = \pi$. One can choose $t_c$ as the time when one wishes the cones to emerge, here we set $t_c/T = 15$. Experimentally that protocol implies changing $\alpha$ after every kick. The Dirac cones form vortices in the $k$-plane with singularities at $(k_c, \alpha_c)$. We demonstrate that the vortices lead to a flip of the molecular orientation during the time evolution even if we time-evolve a generic, experimentally realizable wave packet, populated in one of the bands involved in the crossing.

To this end, we consider a Gaussian wavepacket $(|\phi(t = 0)\rangle \propto e^{-(l-l_0)^2/2\Delta l^2}$ and project this state into the third band with a peak at $k = \pi$ [60]. This furnishes a state which is both localized in angular momentum $l$ and quasimomentum $k$. The occupation in the third band leads to high orientation and alignment signals. Henceforth we time-evolve this wavepacket according to the adiabatic protocol in real space and without approximation of Fourier space conversion.

In Fig. 3 we show the results of this calculation for an initial Gaussian states with width $\Delta l = 0.5$ and peak at $l_0 = 30$ and $l_0 = 0$ respectively. Furthermore, with yellow dots we show the results for a “generic” (not fine-tuned [61]) initial state created by a single laser pulse from $\phi = \delta_{l,0}$. Shown are: (a) the orientation cosine, $\langle \cos(\hat{\theta}) \rangle \equiv \langle \phi(t) | \cos(\hat{\theta}) | \phi(t) \rangle$, (b) the alignment cosine, $\langle \cos^2(\theta) \rangle \equiv \langle \phi(t) | \cos^2(\theta) | \phi(t) \rangle$ and (c) the absolute values squared of the wavefunction components of the “generic” state created by a pulse, $|\langle l | \phi(t) \rangle|^2$. These quantities, expressed through the populations of rotational states and molecular axis distributions in real space, are being routinely measured in gas-phase molecular experiments, e.g., using Coulomb explosion [62] and Raman spectroscopy [63], for (a,b) and (c), respectively.

In the initial time evolution, we observe a stagnant phase with little growth in energy and change in the alignment traces, which is due to the occupation within a specific band. Near the Dirac cones, however, the behavior changes, and we see a change of the orientation and alignment traces. After the cone, the generic behavior predicted at a quantum resonance, namely the linear spread of the wavefunction in angular momentum space is observed. This can also be intuitively understood in terms of a quantum walk in the tight-binding description of the lattice. If we assume that the overlap of the initial wavepacket with the edge states is vanishing, we can expand its Fourier transform in eigenstates with $\phi_k(t = 0) = \sum_n \xi_n \psi_n(k, 0)$, see Eq. (4). The orientation and alignment traces change their behavior at the Dirac cones due to the change of the eigenstates. We note that for a “generic” wave packet which is not prepared exactly in the third band, this phenomenon is still visible, although not as pronounced, depending on the particular setup, see Fig 3. Finally we note that the example we are looking at with $\lesssim 30$ pulses is currently accessible in laboratory, as 24 kicks were used to observe dynamical localization with nitrogen molecules [38].

To summarize, we have demonstrated the possibility to observe nontrivial topological states in periodically kicked molecules. A key step connecting molecular rotational spectroscopy with single-particle models of condensed matter was to introduce quasimomentum, in addition to quasienegries of the Floquet representation. As opposed to topological states in solid-state systems, which usually show up through some macroscopic
observables [2, 64], and topological conical intersections that can alter chemical reaction dynamics [11, 12], the topological states described here can be probed directly by imaging molecular rotational states. In addition to new insights into the topological physics in molecular systems, this paves the way to classify chemically relevant states of molecules using topological invariants. Unlike in many other topological systems, the position of the Dirac cones in molecules can be controlled directly by changing the parametrization of the laser intensities. Furthermore, laser pulses can be applied in a way selectively breaking the symmetries (e.g., time-reversal, but not inversion). The richness of internal degrees of freedom even for diatomic molecules [65] along with the possibilities to coherently control them [44] makes the extensions of the model to arbitrary number of bands with non-Abelian topological invariants possible. M.L. acknowledges support by the European Research Council (ERC) Starting Grant No. 801770 (ANGULON).

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Supplemental material
Multiband topological phases of periodically kicked molecules
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I. TECHNICAL DETAILS

A. Fourier transforms of angular momentum lattices

Here we show that the Fourier transform of the angular momentum lattice provides a viable tool to analyze the Floquet spectra at quantum resonances of a periodically driven molecule. As elaborated in the main text, at a quantum resonance, the time-translational operator takes the form $\hat{U} = e^{-\pi i L^2/N} e^{i l P_1 P_2}$. For odd $N$, the rotational part is periodic with $N$, while for even $N$ the periodicity is $2N$, which becomes evident when looking in the $l$-basis with $\langle l | e^{-\pi i L^2/N} | l' \rangle = e^{-i \pi r(l+1)/N} \delta_{ll'}$. Our approach works for all laser potentials which converge to a constant value, i.e. $V_{l,l'} \approx V_{l+1,l'+1}$, which is in particular true for the combination of a “regular” multi-cycle laser pulse (the $\cos(\hat{\theta})$ term) and a half-cycle laser pulse (the $\cos(\theta)$ term), which can be proven using the Edmonds asymptotic formula for the $3j$-symbols [1]. The asymptotic expressions take the simple form i.e.

$$\langle l'm' | \cos(\theta) | lm \rangle = -\delta_{m'm'} C'_{lm10} C^{10}_{l'010} \delta_{m'm} \delta_{l,l'+1} \delta_{l,l'-1} / 2$$  \hspace{1cm} (S1)

$$\langle l'm' | \cos^2(\theta) | lm \rangle = \delta_{m'm'} \left( \frac{2}{3} C'_{lm20} C^{20}_{l'020} + \frac{1}{3} \delta_{ll'} \right) \delta_{l,l'+1} \delta_{l,l'+2} \delta_{l,l'-2} / 2$$  \hspace{1cm} (S2)

with the usual Clebsch–Gordan coefficients $C^{LM}_{lm} = \langle l', l; m', m | l, L, M \rangle$. To prove this asymptotic, one can use the Edmonds asymptotic formula for $3j$-symbols [2]

$$\langle j_1 m_1 j_2 m_2 | J M \rangle = (-1)^{-j_1+j_2-M} \sqrt{2J+1} \binom{j_1}{m_1} \binom{j_2}{m_2} \binom{J}{M}.$$  \hspace{1cm} (S4)

For Eq. (S1) this leads to

$$C'_{lm10} C^{10}_{l'010} = (-1)^{-m} \sqrt{(2l'+1)(2l+1)} \frac{1}{0 \ m \ l} \frac{1}{0 \ 0 \ l'} \delta_{m,m} \frac{d_{l,l-1}(\theta)}{\sqrt{l_2+l_3+1}} \frac{d_{l',l}(\theta)}{\sqrt{l_2'+l_3'+1}}$$  \hspace{1cm} (S3)

where $d_{m,m}(\theta)$ is the Wigner function and $3j$-symbols are related to the Clebsch-Gordan coefficients [2]

$$\langle j_1 m_1 j_2 m_2 | J M \rangle = (-1)^{-j_1+j_2-M} \sqrt{2J+1} \binom{j_1}{m_1} \binom{j_2}{m_2} \binom{J}{M}.$$  \hspace{1cm} (S4)

For Eq. (S2) this derivation proceeds analogously, with the difference that $d_{0,\pm2}(\tau/2)d_{0,\mp2}(\tau/2) = 3/8$ and $d_{0,\pm2}(\tau/2)d_{0,\pm2}(\tau/2) = 3/8$ and $d_{0,0}(\tau/2)d_{0,0}(\tau/2) = 1/4$, which in total leads to the desired result. For the projection of angular momentum $m = 0$ these coefficients converge fast, see Fig. S1, and the overall behavior of the system is very well described by an approximation where we assume a constant value. In this work we only consider $m = 0$, but for other $m$ the approximations work out as well when considering larger $l$. The $N$-periodicity allows us to define a Fourier transform which approximates the periodic behavior for $l, l' \gg 0$. Any operator $\hat{A}$ with that periodicity, and the effective Hamiltonian in particular, follows

$$\langle l' | \hat{A} | l \rangle \approx \langle l' + N | \hat{A} | l + N \rangle$$  \hspace{1cm} (S6)

for $l, l' \gg 0$. Let us parametrize each $l', l$ by $l = n \cdot N + i$, $l' = n' \cdot N + j$ with $n, n' \in \mathbb{N}_0, i, j \in \{0, 1, \ldots, N\}$. Then,

$$A_{l'l} = A_{i,j}(n', n) = A_{i,j}(n' - n) = A_{i,j}(\Delta n)$$  \hspace{1cm} (S7)
Figure S1. Convergence of the hopping elements for \( l, l' \gg 0 \) to a constant value. In (a) we show the element which is relevant for the \( \cos(\theta) \) potential and in (b) for the \( \cos^2(\theta) \). In (c) we show the real part of the time-translation operator, \( \hat{U} = e^{-\frac{\pi}{2} L^2/k} e^{i \hat{V}(\theta)} \), with \( N = 3, P = 2.5 \) and \( \alpha = \pi/3 \) for the potential \( V(\theta) = P \left( \cos^2(\alpha) \cos^2(\theta) + \sin^2(\alpha) \cos(\theta) \right) \) as in the rest of the letter. We marked the unit-cell size \( N = 3 \) by gray lines.

i.e. A only depends on the off-diagonal index \( \Delta n \) because of the periodicity. The Fourier transform from \( l \)- to \( k \)-space then becomes (for a dimensionless momentum variable \( k \in [0, 2\pi] \))

\[
A_{ij}(k', k) = \sum_{n', n} e^{-i(n' - n)k} A_{ij}(n', n) = \sum_{\Delta n} e^{-i(n'k - nk)/2 + \Delta n(k + k')/2} A_{ij}(\Delta n) = \delta_{kk'} \sum_{\Delta n} e^{-i\Delta n - k} A_{ij}(\Delta n),
\]

\[
(i.e. \quad A_{ij}(k) = \sum_{\Delta n} e^{-i\Delta n - k} A_{ij}(\Delta n) \quad \text{and correspondingly} \quad A_{ij}(\Delta n) = \frac{1}{2\pi} \int_0^{2\pi} A_{ij}(k) dk. \quad \text{(S8)}
\]

where we used \( \bar{n} = n + n' \), \( \Delta n = n'-n \) and \( A_{ij}(n', n) = A_{ij}(\Delta n) \), which shows that operator \( \hat{A} \) conserves momentum \( k \). Since the Fourier transform is only well defined for an infinite system, for a finite system this implies practically an infrared cutoff in momentum which is defined by the system size. When calculating the Fourier transform of an operator, one needs to choose a unit-cell in the middle of the lattice to avoid edge effects. Practically, we choose \( A_{ij}(\Delta n) = A_{N, n_0 + i, N, (n_0 + \Delta n) + j} \) with a large enough \( n_0 \) to ensure convergence. For an infinite and perfectly translationally invariant system, we would sum over all \( \Delta n \in \mathbb{Z} \). However, here we need to choose \( -n_0 \ll \Delta n \ll n_0 \). For small to moderate \( P \), only few off-diagonals suffice to achieve high agreement with the “real-space” diagonalization. A state transforms with

\[
f_i(k) = \sum_{\Delta n} e^{-i\Delta n - k} f(i + N(n_0 + \Delta n)), \quad f(i + N(n_0 + \Delta n)) = \frac{1}{2\pi} \int_0^{2\pi} f_i(k) e^{i\Delta n - k} dk. \quad \text{(S9)}
\]

The Fourier transform of the operators at \( l', l \gg 0 \) in (S1) and (S2) is given by

\[
(cos(\theta))_{ij}(k) = \frac{1}{2} \begin{pmatrix}
0 & 1 & \cdots & e^{-ik} \\
1 & 0 & 1 & \cdots \\
\vdots & \vdots & \ddots & \vdots \\
e^{+ik} & \cdots & 1 & 0
\end{pmatrix}, \quad \text{N×N Matrix} \quad \text{(S10)}
\]
and \((\cos^2(\hat{\theta}))_{ij}(k) = \frac{1}{2} \left( \begin{array}{cccccc}
1 & 0 & 1/2 & \cdots & e^{-ik/2} & 0 \\
0 & 1 & 0 & 1/2 & \cdots & e^{-ik/2} \\
1/2 & 0 & 1 & 0 & 1/2 & \cdots \\
\vdots & \ddots & \ddots & \ddots & \ddots \ \\
1/2 & 0 & 1 & 0 & 1/2 & \cdots \\
e^{+ik/2} & 0 & 1 & 0 & 1/2 & \cdots \\
e^{-ik/2} & 0 & 1 & 0 & 1/2 & \cdots \\
\end{array} \right) \).

To calculate \(e^{i\hat{V}}\) it is sufficient to diagonalize \(V_{ij}(k)\) and express the exponential in terms of its eigenstates \(v_n(k)\) and eigenvalues \(\lambda_n(k)\), i.e. \(e^{i\hat{V}(k)} = \sum_n e^{i\lambda_n(k)} (v_n(k) \otimes v_n^\dagger(k))\).

### B. Symmetries

The rotational phases, \(\phi_l = e^{-\pi l(l+1)/N}\), give rise to reflection symmetry for all \(N\) with translationally-invariant potentials (i.e. in our case for \(l', l \gg 0\)). For an odd \(N\), the reflection center of the first unit cell is \(n_c = (N+1)/2 \in \mathbb{N}\). Then, for \(0 \leq j < n_c\) we have \(\phi_{n_c+j} = \phi_{n_c-j}\) (for even \(N\), the unit-cell is \(2N\) and \(n_c = N\) with \(\phi_{n_c+j+1} = \phi_{n_c-j}\)). If we limit ourselves to a finite Hilbert space with maximum \(l_{\text{max}}\), we can write the symmetry in the \(l\)-basis as

\[
\mathcal{R} = \begin{pmatrix}
\cdots & 0 & 1 \\
0 & 1 & 0 \\
1 & 0 & \cdots \\
\end{pmatrix}
\]

with \(\mathcal{R}'\mathcal{R} = \mathcal{R}^2 = 1\). It commutes with the time-translation operator, \([U, \mathcal{R}] = 0\). In terms of the effective Hamiltonian, \(\hat{H} = i \log \hat{U}\), this symmetry reads in \(k\)-space \(\mathcal{R} \hat{H}_k = \hat{H}_{-k} \mathcal{R}\) with

\[
\mathcal{R} = \begin{pmatrix}
\cdots & 0 & 1 \\
0 & 1 & 0 \\
1 & 0 & \cdots \\
\end{pmatrix} \quad \text{and the parity operator with} \quad \mathcal{P} \phi_{-k} = \phi_{k}.
\]

For the one-kick system, shifting the position of the kick does not change the model, i.e.

\[
U(\beta) = e^{-iL^2\pi(1-\beta)/N}e^{i|V_1|}\quad \sim \quad U = e^{-iL^2\pi/|V_1|}
\]

where the \(\sim\) implies that the two operators have the same spectrum up to a constant for any \(0 \leq \beta \leq 1\). However, as we have seen above, a finite \(\beta\) leads to different \(N\)-periodicity. Let us write \(\beta = \frac{a}{b}\) with \(a, b \in \mathbb{N}, a < b\). Then, for incommensurable \(a, b \cdot N\), the periodicity of the operator will be \(b \cdot N\) (correspondingly, for commensurable \(a\) and \(N \cdot b\) it will be the lowest common denominator). The equivalence of the spectra then leads to \(bN\) bands in place of \(N\) bands which compensates for the larger unit-cell. This is a well-known phenomenon commonly known as “band folding” in solid-state physics.

In addition to that, we find a time-reflection symmetry present for all models with one kick in the Floquet operator (where we use that our laser potentials follow \(V^* = V = V^T\)). More specifically, the second kick (for instance, \(U_{2\text{-kick}} = e^{-iL^2\pi/|V_1|}e^{i|V_1|}\)) would break the time-reflection invariance. The time-reflection invariance is proportional to unity times conjugation when the pulse is at the center, i.e. \(\beta = 1/2\). In that gauge, we find

\[
\langle l'|U^*(1/2)U(1/2)|l \rangle = \sum_{l''} \langle l'|e^{+i\pi(l' + 1)/2N}e^{-i\pi l''(l'+1)/2N}e^{-i\pi l''(l'+1)/2N}e^{-i\pi l''(l'+1)/2N}\rangle e^{i\pi l''(l'+1)/2N}e^{-i\pi l''(l'+1)/2N} = \delta_{ll'}
\]
which is equivalent to the time-reflection symmetry of the effective Hamiltonian $H(\beta) = H^*(\beta)$. To construct $T$ for arbitrary gauges $\beta$, we only need to bring $U$ (or $H$, respectively) back to the $\beta = 1/2$ gauge. This is accomplished by the gauge transformation $U(\beta)$ with

$$U(\beta) = e^{-iL^2\pi(2\beta-1)/(2N)} \quad \text{with} \quad U(\beta)U(\beta) = U(1/2). \quad (S16)$$

The (anti-unitary) time-reflection operator then takes the form $T(\beta) = U(\beta)C U(\beta)^\dagger$ with conjugation $C$. The time-translation operator then fulfills $T(\beta)U(\beta) = U(\beta)T(\beta)$, while the Hamiltonian $[H(\beta), T(\beta)] = 0$. In $k$-space, this leads to

$$T(\beta)H_k(\beta) = H_k^*(\beta)T(\beta). \quad (S17)$$

C. $N = 3$ case: Dirac cones

For $N = 3$, the Fourier transform of the laser potential becomes

$$V(k) = \frac{P_1}{2} \begin{pmatrix} 1 & e^{-ik}/2 & e^{-ik}/2 \\ e^{ik}/2 & 1 & e^{-ik}/2 \\ e^{ik}/2 & e^{-ik}/2 & 1 \end{pmatrix} + \frac{P_2}{2} \begin{pmatrix} 0 & 1 & e^{-ik} \\ 1 & 0 & 1 \\ e^{ik} & 1 & 0 \end{pmatrix}$$

$$= \frac{1}{2} \begin{pmatrix} P_1 & P_1e^{-ik}/2 + P_2/2 & P_1e^{-ik}/2 + P_2 \\ P_1e^{ik}/2 + P_2 & P_1/2 + P_2e^{-ik} \\ P_1e^{ik}/2 + P_2 & P_1e^{-ik}/2 + P_2/2 \end{pmatrix}. \quad (S18)$$

The reflection symmetry $1 \leftrightarrow 3, k \leftrightarrow -k$ is clearly visible here. In Fig. S2 we show a comparison of the diagonalization in $l$-space and $k$-space. For small to intermediate coupling ($P \sim 5$) the agreement is very good. If we look at the smallest energy difference $d(P_1, P_2) = \min_{n,m \in \{1,2,3\}} (|\epsilon_n - \epsilon_m|_{2\pi})$ with the $2\pi$-periodic distance, i.e.

$$|x - x'|_{2\pi} = \min(|x - x'|, |x - x' + 2\pi|, |x - x' - 2\pi|), \quad (S19)$$
II. EXPERIMENTAL REALIZATIONS OF DIRAC CONES

The origin of this behavior can be explained by analyzing the eigenstates of the Hamiltonian. At each moment of time, the eigenstates $\psi(k, \alpha) \in \mathbb{C}^3$ can be obtained by

$$H(k, \alpha(t))\psi(k, \alpha(t)) = \epsilon(k, \alpha(t))\psi(k, \alpha(t)) \quad (S21)$$

with $\alpha(t) = t \cdot (\alpha_c/t_c)$ and the critical value $\alpha_c$ with the Dirac cone, see Fig. S5. In Fig. S6, we recognize the Dirac cone as a vortex in the expectation value of the eigenstate $\langle \psi_k | \cos^2(\theta) | \psi_k \rangle$. The Dirac cone causes a drastic change in the orientation and alignment of the eigenstates. A state $\phi(t)$ which consists of a superposition of different momentum eigenstates mimics their evolution accordingly. The drastic change is only observed for states with large occupation at $k = \pi$, since that is the position of the Dirac cone at $t = t_c$. The states for the time-evolution in the main text are obtained as follows. We begin with a Gaussian state in angular momentum space, $\phi_0 \propto e^{-(t-t_0)^2/2\Delta^2(t)}$ and project it into the third band, i.e. $|\phi_0\rangle \rightarrow \sum_{i \in \mathbb{N}_3} |\psi_i\rangle\langle\psi_i|\phi_0\rangle$ with all Floquet states $i \in \mathbb{N}_3$ of the third band. Then, in order to

$Figure S3. Smallest quasienergy differences (i.e. smallest band gap) for $k = \pi$ (left figure) and $k = 0$ (right figure). For small to intermediate couplings the only way to achieve gap closings is with $P_1 \neq 0, P_2 \neq 0$. 

then we observe that the gap closings occur for $k = 0$ and $k = \pi$ on straight lines, see Fig. S3. Note that the linearity of the gap closings can be turned into non linear curves using multiple pulse schemes and more complicated potentials. Furthermore, we mention in the main text that the topological charge of the Dirac cones can be evaluated.
Figure S4. The three energy bands, $\epsilon_n(k, \alpha)$ at $P = 2.5$, from (a) lowest to (c) highest. The dashed lines mark exemplary integration contours around the Dirac cones, which provide a way to evaluate the topological charge of the cones, see text. The color of the lines denote the result of the integration with $+\pi$ (red lines) and $-\pi$ (blue lines).

guarantee a peak at $k = \pi$, we convolve this state with a Gaussian distribution centered at $k_0 = \pi$, that is

$$\phi_k \propto \int_0^{2\pi} e^{-((k-k_0)/2\Delta k)^2} (\phi_k \cdot \psi_k^*) \psi_k \, dk$$

(S22)

where we choose $\Delta k = 0.5$. Since this convolution removes overlap with the original Gaussian distribution in $l$-space, it can lead to a delocalization of the former localized state. Clearly, this is a very simple procedure and choosing a more complicated initial state with a well-defined momentum distribution is also possible.

In Fig. S7 we show the time evolution of the states discussed in the main text. In the beginning of the time evolution, the states do not spread but remain more or less localized, since only a single band is occupied. After crossing the Dirac cone, more bands are occupied (this is a non-adiabatic effect) and the wavepacket spreads. However, the change of the alignment/orientation measures is due to the change of the eigenstate, not due to the change of bands, as explained in Fig. S6. The Dirac cone marks the position where these measures suddenly change, and henceforth also the states which depend on them. This implies that by tuning the position of the Dirac cone in terms of the laser intensities, we can also tune when this changes happen for an arbitrary wave function.

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Figure S5. The quench protocol of the main text and the corresponding changes of the spectrum in time. For the chosen parameters, $P_1 = 1.5, P_2 = 1.6$, the critical value of the parameter is $\alpha_c \approx 0.6221$. The parametrization reads $\alpha(t) = t \left( \alpha_c / t_c \right)$, where we choose $t_c = 15$. As for the spectrum in the main text, for $\alpha > \alpha_c$ two edge states emerge in the middle of the two upper bands. They can only be removed by a second cone in between the two bands.

Figure S6. The orientation and alignment signals of the instantaneous eigenstates of the Hamiltonian at a given moment of time. The panels (a),(b),(c) correspond to the first, second and third band, respectively. In the first row we show the expectation value of $\cos(\theta)$ and in the second of $\cos^2(\theta)$. The red dashed line marks the occurrence of the Dirac cone. The Dirac cone at $t_c = 15$ leads to a vortex-like structure in the phases of the eigenstates, which translates into a similar vortex in orientation/alignment signals. For $k = \pi$, the change is most drastic, e.g. $\langle \psi_{k=\pi} | \cos(\theta) | \phi_{k=\pi} \rangle$ changes sign after passing $t_c$. This figure explains the pronounced changes in the signals of Gaussian wavepackets peaked at $k = \pi$, as shown in the main text, when assuming a quasi-adiabatic time evolution. Note that the symmetry in quasimomentum $k$ is due to the inversion symmetry of the Hamiltonian and will persist after breaking the time-reversal symmetry (for instance, by introducing additional kicks). The change of the behavior of the eigenstates can be explained using the inversion quantum number $n_i = \pm 1$, given by the eigenvalues of the inversion operator defined earlier for inversion-invariant momenta $k = \pi$ and $k = 0$. At these momenta, the Hamiltonian becomes block-diagonal. The Dirac cones furnish an exchange of a $n = +1$ state with the $n = -1$ between the bands while maintaining the band structure. This exchange of the states leads to a drastic change of the alignment/orientation signal of the whole band after the cone.
Figure S7. Time evolution of the states discussed in the main text, also see (S22). (a) and (b) show Gaussian states with $\Delta l = \Delta k = 0.5$ with (a) $l_0 = 30$ and (b) $l_0 = 0$. (c) shows a “generic” state created by a laser pulse starting from $l = 0$, $|\phi\rangle = e^{-i\Delta t \hat{L}^2} e^{iV(P_1, P_2)} |l = 0\rangle$, with $\Delta t \approx 0.911, P_1 \approx 6.67, P_2 \approx 2.67$. These parameters ensure that the state has a large overlap with the third band and is sufficiently occupied around $k = \pi$. We observe that in all three exemplary time evolutions the behavior changes drastically in the vicinity of the Dirac cone. This can be explained by the change of the state due to the band crossing and its spread over different bands, see Fig. S6.