Topological Charges 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 topological charges 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 us to define quasimomenta 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. Within the synthetic dimension of the laser strength, we discover Dirac cones with topological charges. These Dirac cones, topologically protected by reflection and time-reversal symmetry, are reminiscent of (although not equivalent to) that seen in graphene. They—and the corresponding edge states—are broadly tunable by adjusting the laser strength 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.

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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]. These phenomena are characterized by *topological charges*—a type of a quantum number that describes the topology of the system. These topological phases show unique properties, such as quantized transport and the bulk-boundary correspondence [2]. The field of topological phases has greatly expanded since the works of Thouless [3] and Haldane [4,5], leading to discoveries of several new phases, 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 use a novel approach and allow us to directly bridge the ideas of symmetry-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 us to probe the nontrivial topology of

their rotational states. This allows us to apply the vocabulary and theorems developed for topological materials in the realm of chemical physics, and to potentially engineer topologically protected molecular states with new chemical applications. 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 [48], we can write the Hamiltonian (in units of $\hbar \equiv 1$) as follows [40,60]:

$$\hat{H}_{\mathrm{mol}}(t) = B\hat{\mathbf{L}}^2 - \underbrace{[P_1\cos^2(\hat{\theta}) + P_2\cos(\hat{\theta})]}_{\equiv \hat{V}(P_1,P_2)} \sum_{q=0}^{\infty} \delta(t - qT). \quad (1)$$

Here, $B = \pi/\tau_B$ is the molecular rotational constant with τ_B the rotational period, $\hat{\mathbf{L}}$ is the angular momentum operator, with centrifugal distortion neglected [61]. The laser pulses are defined by their strengths, P_1, P_2, and the time period between them, T. $\hat{\theta}$ gives the angle between the linear polarization of both lasers and the molecular axis. Unlike most other models taking into account either the $\cos^2(\hat{\theta})$ term ("regular" multicycle infrared pulses) or the $\cos(\hat{\theta})$ term (terahertz half- or few-cycle pulses [55,56,60,62,63]), here we include both, which allows us to create tunable Dirac cones, not observable with either one of the terms [48]. For simplicity, we combined the two laser pulses in one potential, but the same behavior can be observed for two consecutive pulses. Both kind of pulses have been realized independently in experiments and used for orientation and alignment of molecules [41–43,64–66].

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 Letter, in order to achieve a banded system, we focus on the resonant regime [35,67], i.e., $T = \tau_B/N$. The time-translation operator of the Hamiltonian defined in Eq. (1) after one driving period takes the form

$$\hat{U} = \underbrace{e^{-\pi i \hat{\mathbf{L}}^2/N}}_{\text{free rotation}} \underbrace{e^{i\hat{V}(P_1,P_2)}}_{\text{kick}}. \quad (2)$$

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, ϵ_n [68,69]. 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 if N is odd (for even N the periodicity is $2N$ and there are $2N$ bands, see Ref. [48]), which is a result of the N periodicity of the $e^{-\pi i\hat{\mathbf{L}}^2/N}$ operator.

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, $H_{l,l'} \approx H_{l+N,l'+N}$ for $l, l' \gg 0$, with $H_{l,l'} = \langle l|\hat{H}|l'\rangle$, cf. Eq. (1) [48]. This allows us 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 $l, l' \gg 0$, which, however, suffices for the purposes of our proposal. Let us assume that $l \in \mathbb{Z}$, then, the

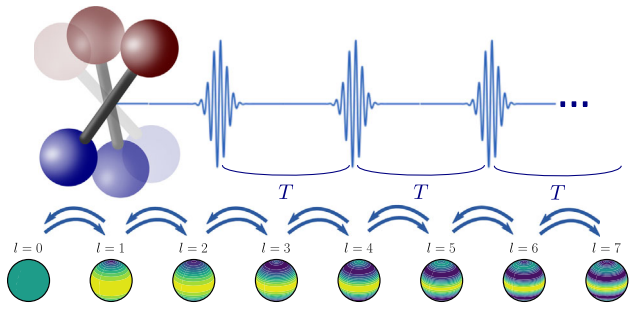

FIG. 1. Illustration of the angular momentum lattice with the spherical harmonics of the molecule and the hopping between different lattice sites due to the periodic laser pulses, cf. Eq. (1). For $l \gg 0$, the hopping terms converge to a constant and the lattice becomes translationally invariant. The hopping terms are complex numbers given by the time-translation operator (see Fig. S4 in Supplemental Material [48]).

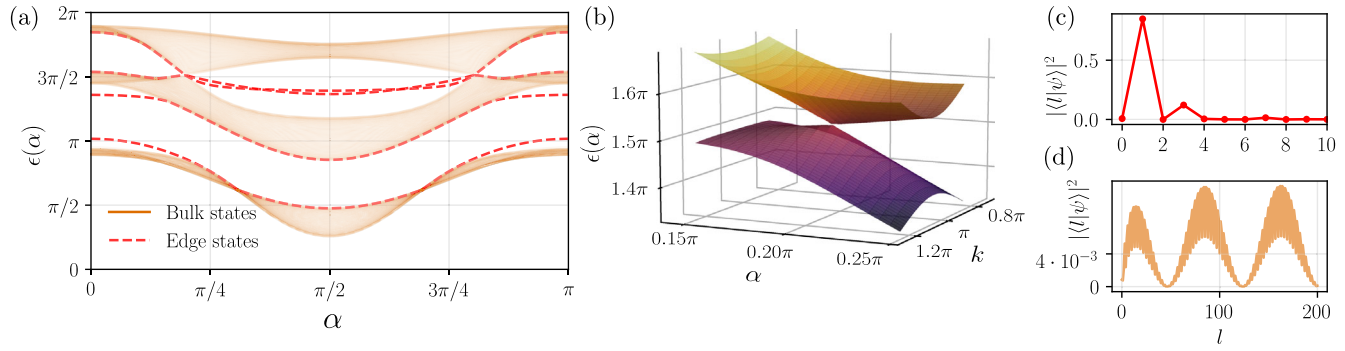

FIG. 2. Results of exact diagonalization for $N = 3$ and $P = 2.5$, see Eq. (5). (a) The full spectrum with the bulk states shown in orange and the edge states in red. Two linearly dispersing edge states (one for each edge) connect the Dirac cones. (b) The quasienergies ϵ_k near the Dirac cone, shown as a function of k and α. 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, unless there is a merging transition where a positively charged cone annihilates a negatively charged one. (c) The absolute value of the wave function of the edge state. (d) A generic bulk state.

periodicity of the lattice allows us to define the Fourier transform with quasimomentum $k \in [0, 2\pi)$ by

$$\underbrace{H_{ij}(k)}_{N\times N \text{ matrix}} = \sum_{\Delta n} e^{-i\Delta n\cdot k} H_{ij}(\Delta n), \tag{3}$$

with $H_{ij}(\Delta n) = H_{i,N\cdot\Delta n+j}$. (In practice, we need to choose a cutoff for a finite system, see Ref. [48]. 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

$$f_i(k) = \sum_{\Delta n} e^{-i\Delta n\cdot k} f(i + N\Delta n). \tag{4}$$

Correspondingly, $H(k)\psi_n(k) = \epsilon_n(k)\psi_n(k)$ where k plays the role of quasimomentum of the angular momentum lattice and $\psi_n(k) \in \mathbb{C}^N$ [note that $H(k)$ and other operators are $N \times N$ matrices in k space].

Since there are N bands, the Hamiltonian can be written as $H(k) = a(k)\mathbb{I}_N + \sum_i d_i(k)\,\lambda_i$ 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 $a(k) \in \mathbb{R}$, $\mathbf{d}(k) \in \mathbb{R}^{N^2-1}$, see Ref. [70]. 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 [71,72], 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 strength parameters:

$$P_1(\alpha) = P\cos^2(\alpha), \qquad P_2(\alpha) = P\sin^2(\alpha), \tag{5}$$

in terms of the offset parameter $\alpha \in [0, 2\pi)$, which serves as an effective second dimension in addition to quasimomentum 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 $m = 0$. Figure 2 shows the results of exact diagonalization. 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 [48], i.e.,

$$\mathcal{T}H_k = H_{-k}^*\mathcal{T} \quad \text{and} \quad \mathcal{R}H_k = H_{-k}\mathcal{R}. \tag{6}$$

This implies that for the $k \to -k$ invariant points, i.e., $k = 0$ or $k = \pi$, the Hamiltonian commutes with $\mathcal{T}$ and $\mathcal{R}$. From the tenfold classification with crystal symmetries the class is $AI_+\mathcal{R}_+$ [73]. The topological analysis of the two Dirac cones proceeds as follows. We combine the two variables into a vector $\mathbf{k} = (k, \alpha)$, and compute the Berry connection, $\mathcal{A}_n(\mathbf{k}) = i\langle\psi_n(\mathbf{k})|\nabla_{\mathbf{k}}\psi_n(\mathbf{k})\rangle$, which is well defined if we choose a gauge such that the eigenstates form a smooth manifold. Numerically evaluating the Berry phase [74] around one of these cones in the second or third band results in $\int_\gamma \mathcal{A}(\mathbf{k})_n d\mathbf{k} = \pm\pi$ thus proving the topological nature of these cones [75]. 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 (or upper [76]) end of the lattice. Topologically nonprotected 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 α of Eq. (5) as

$$\alpha(t) = t \cdot (\alpha_c/t_c), \tag{7}$$

where α_c denotes the critical value of α 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 α after every kick. The Dirac cones form vortices in the $\mathbf{k}$ plane with singularities at (k_c, α_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 wave packet $\langle l|\phi(t=0)\rangle \propto e^{-(l-l_0)^2/2\Delta l^2}$ and project this state into the third band with a peak at $k = \pi$ [77]. 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 wave packet 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" initial state created by a single laser pulse from $\phi = \delta_{l,0}$. Generic implies that we are choosing a state which can be created by one laser pulse starting from $l = 0$. Evidently, to see a difference in the alignment and orientation signal, one needs to start from a state which is somewhat aligned or oriented [78]. 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 $\langle\cos^2(\hat{\theta})\rangle \equiv \langle\phi(t)|\cos^2(\hat{\theta})|\phi(t)\rangle$, and (c) the absolute values squared of the wave function 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 [57] and Raman spectroscopy [79], for (a),(b), and (c), respectively.

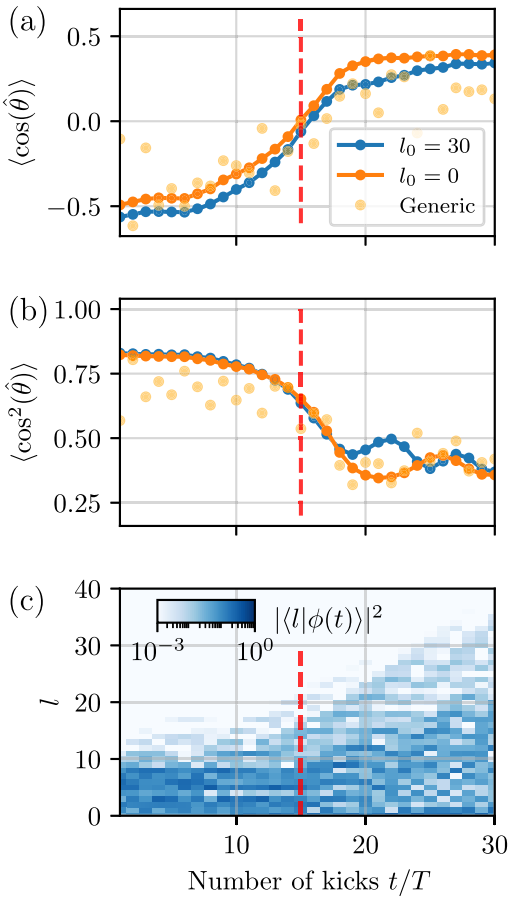

FIG. 3. Time evolution of three different molecular states initially prepared in the third band of the spectrum peaked at $k = \pi$, see Fig. 2(a), which are evolved through the Dirac cone at $t_c = 15$, cf. Eq. (7). To demonstrate that the phenomenon is generic we show a Gaussian state peaked at $l_0 = 30$ (orange), $l_0 = 0$ (blue), and a generic state prepared with one laser pulse from $l = 0$ (dotted). As a function of the number of laser kicks are shown (a) the orientation cosine $\langle\cos(\hat{\theta})\rangle$, (b) the alignment cosine $\langle\cos^2(\hat{\theta})\rangle$, and (c) the absolute values squared of the wave function components $|\langle l|\phi(t)\rangle|^2$ of the generic state. The dynamics changes drastically in the vicinity of the Dirac cone, marked by vertical dashed lines (red), see the text. The reason for this is a monopole at $k = \pi$, $\alpha = \alpha_c$ which changes the nature of the eigenstates.

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 wave function 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 wave packet 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 topological charges 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 quasienergies of the Floquet representation. As opposed to topological states in solid-state systems, which usually show up through some macroscopic observables [2,80], and topological conical intersections that can alter chemical reaction dynamics [11,12], the topological charges 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 engineer 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 strengths. Furthermore, laser pulses can be applied in a way selectively breaking the symmetries (e.g., time reversal). The richness of internal degrees of freedom even for diatomic molecules [81] along with the possibilities to coherently control them [44] makes the extensions of the model to higher dimensions possible. Using elliptically polarized laser pulses the space of rotations for an asymmetric molecule becomes three dimensional (in the quantum numbers l, m, k) and a hopping in all the three dimensions can be induced [82]. Applying pulses which are in resonance with the rotational periods, multiband and possibly non-Abelian physics in more than one physical dimension could be probed. Last, previous works indicate that immersing rotating molecules into superfluid helium [83] might generate an interacting topological system with non-Abelian invariants [84].

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[75] Another possibility is to calculate the mirror invariants, which can be computed from the difference in the number of reflection-symmetric eigenstates at $k = 0$ (or $k = \pi$) before and after the cone.

[76] From the rigid rotor approximation, the lattice in l is assumed to be half-infinite. However, that is only an approximation, see Refs. [61,37]. The centrifugal distortion leads to an effective upper bound $l_{\max}$, where another edge state appears.

[77] Quasimomentum $k = \pi$ implies that the sign of the matrix elements changes every three angular momentum sites.

[78] We choose $|\phi\rangle = e^{-i\Delta t\hat{L}^2} e^{iV(P_1,P_2)}|l = 0\rangle$ with $\Delta t \approx 0.911$, $P_1 \approx 6.67$, $P_2 \approx 2.67$. However, any other state with finite occupation in one band around $k = \pi$ can be used.

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