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Quasiperiodicity and 2D Topology in 1D Charge Ordered Materials

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It has recently been argued that individual 1D quasicrystals can be ascribed 2D topological quantum numbers and a corresponding set of topologically protected edge modes. Here, we demonstrate the equivalence of such 1D quasicrystals to a mean-field treatment of incommensurate charge order in 1D materials. Using the fractal nature of the spectrum of commensurate charge-ordered states we consider incommensurate order as a limiting case of commensurate orders. We show that their topological properties arise from a 2D parameter space spanned by both phase and wave vector, bringing the observation of 2D edge modes in line with the standard classification of topological order. The equivalence also provides a set of real-world quasiperiodic materials which can be readily experimentally examined. We propose an experimental test of both the quasicrystalline and topological character of these systems in the form of a quantized adiabatic particle transport upon dragging the charge-ordered state.

The second half of the twentieth century saw two revolutions in condensed matter physics. The first began with the mathematical observation that solids need not involve periodic repetitions of unit cells, but can instead involve aperiodic tilings of two or more inequivalent cells [1]. Experimental verification in the form of a ‘quasicrystalline’ Al-Mn alloy followed shortly afterwards [2].

The second revolution regarded the observation and subsequent theoretical characterization of the quantum Hall effect [3, 4]. This prompted the realization that phase transitions cannot all be characterized solely in terms of the continuous symmetries they break. It lead to a new, topological, classification scheme of all non-interacting fermion theories according to discrete symmetries, in what has come to be known as the ‘Tenfold Way’ [5, 6].

Recently these two cornerstones of modern condensed matter physics have been shown to be mathematically linked through the ubiquitous Harper equation [7]. This equation has long been known to govern electrons on a 2D lattice in the presence of a magnetic field, i.e. the quantum Hall effect [8]. It was also recently shown to provide a description of 1D quasicrystals when used to describe electrons in a 1D lattice with an incommensurate periodically modulated on-site potential [9]. Based on the equivalence of the underlying mathematical structures of these two systems, quantized transport properties analogous to the quantum Hall conductance were predicted for the quasicrystal, and subsequently observed in an experimental realization using optical waveguides [10].

Quasicrystals—Quasicrystals are defined to be quasiperiodic tilings of two (or more) inequivalent unit cells [1]. A quasiperiodic tiling in 1D can be generated from the projection of a regular 2D crystal [1]. As shown in Figure 1, a straight line can be drawn in a square 2D lattice, in such a way that it hits exactly one lattice point. A second line is then drawn parallel to the first, going through the opposite corner of the unit cell. Whenever a point of the 2D lattice falls between the lines, its projection onto the first line forms a site of the 1D quasicrystalline lattice. The projected lattice consists of precisely two unit cells, of different sizes. These appear to be randomly tiled, in a never-repeating
Incommensurate
2\pi/Q = 2a
Commensurate
Quasicrystal

FIG. 1. **Main:** generating a 1D quasicrystal by projecting a 2D lattice. A pair of parallel lines are overlaid on the square lattice, in such a way that they hit only a single pair of lattice points on opposing corners of one of the square unit cells. The lattice points between the lines are then projected, and form the sites of the 1D quasicrystal. The inequivalent cells in the quasicrystalline lattice are colored red and blue for clarity. **Top Inset:** generating an incommensurate charge-ordered state with period \(2\pi/Q\) on an atomic lattice with spacing \(a\), by a similar projection method. In this case the sequence of atomic sites and maxima of the charge density modulation (red balls and blue tick marks) is quasiperiodic. **Bottom Inset:** the same method also trivially generates commensurate charge order, when the angle between the two lines is zero.

pattern, but are in fact perfectly predictable: the type of unit cell at a given location is known with certainty owing to the periodicity of the 2D parent lattice. This is the definition of quasiperiodicity adopted here.

A remnant of the 2D projection underlying the 1D quasicrystal can be seen in its diffraction pattern, which is generated by two different reciprocal lattice vectors \(1\). This argument was recently employed to explain the observation of an integer number of edge states in 1D quasicrystals realized in optical waveguide arrays \(10\). The number of edge states is a direct measure of the Chern number of a system, which depends only on the system’s topological character \(4\). According to the accepted classification of fermion systems with no assumed symmetries, Chern numbers that can take any integer value, as in the waveguide experiments, exist in systems with even spatial dimensions \(5\). Odd-dimensional systems have only trivial topological phases, without edge modes \(10\) \(17\).

An alternative explanation of the seemingly paradoxical presence of 2D Chern numbers in 1D quasicrystals builds on the assertion that the waveguide experiments involve two degrees of freedom rather than one \(10\). The additional parameter relates distinct but locally isomorphic quasicrystals, where two quasicrystals are defined to be locally isomorphic if and only if every finite sequence of unit cells which appears in one also appears in the other \(1\). The experimentally observed edge states and their related quantized transport properties are then argued to relate to the entire 2D family of quasicrystals, rather than to any one of its 1D members \(10\).

**Charge Order**—The argument about the effective dimensionality and topological character of quasicrystals can be straightforwardly reformulated in terms of properties of charge-ordered (quasi-)1D materials. In such systems chains of atoms spontaneously develop a modulation in their electronic density that does not coincide with the periodicity of the underlying ionic lattice. They include well-studied materials like NbSe\(_3\), KCP, and TTF-TCNQ, which all contain weakly coupled 1D chain structures \(18\)\(22\).

The connection with quasicrystals can be made using a simple projection method. Consider a 1D atomic lattice with spacing \(a\) and a sinusoidal modulation of charge density with period \(2\pi/Q \neq a\), as indicated in the inset of Figure 1. For incommensurate charge order, the sequence of atomic sites and maxima in the charge modulation along the chain form a seemingly random, never-repeating, pattern. It can be easily seen however that the pattern is perfectly predictable as it arises from the projection of a second 1D lattice with lattice spacing \(na\), where \(n\) is the lowest integer number larger than \(2\pi/Qa\). This second lattice can always be rotated to a point where the projections of its lattice sites onto the original line coincide with the maxima of the charge modulations (see Figure 1). The sequence of sites and charge maxima in the original lattice is therefore quasiperiodic. The combined system can be interpreted as a quasicrystal, which could in principle be observed in scanning tunneling microscopy experiments. By continuously shifting the charge order with respect to the atomic lattice, a continuous set of locally isomorphic quasicrystals is generated.

The emergence of charge-ordered states can be most easily understood in a model of spinless fermions hopping on a 1D lattice, in the presence of nearest-neighbor density-density interactions (strength \(h\)) \(22\). Such a 1D system is always unstable towards a spontaneous modulation of its local electronic density. This can be described at the mean-field level by introducing the expectation value \(\Delta_Q(h) = 2h \sum_k \langle \hat{c}^\dagger_{k+Q} \hat{c}_k \rangle\), where \(Q\) is a wave vector connecting the two points on the Fermi surface of the 1D band structure. Using this decoupling, the mean-field Hamiltonian can be written as:

\[
\hat{H} = \sum_{0 \leq k < 2\pi/a} \left\{ \frac{1}{2} \epsilon_k \hat{c}^\dagger_k \hat{c}_k + \Delta_Q \hat{c}^\dagger_{k+Q} \hat{c}_k + H.c. \right\}
\]  

where \(\epsilon_k = 2t (1 + \cos (ka)) - \mu\) describes the bare band structure resulting from the hopping integral \(t\) and chemical potential \(\mu\), and the charge density order parameter can be written as \(\Delta_Q = |\Delta_Q| \exp (i\theta)\). If the chemical potential is tuned to a rational fraction \(p/q\) of the bare bandwidth \(4t\), the charge order in the ground state of the full mean-field Hamiltonian in Equation (1) will have period \(qa\). Working in a reduced Brillouin zone of length...
2π/a, the Hamiltonian can then be rewritten as:

\[
\hat{H} = \sum_{0 \leq k < 2\pi/a} \left( \hat{c}_{k+Q}^\dagger, \hat{c}_{k+2Q}^\dagger \ldots \hat{c}_{k+qQ}^\dagger \right) H_k \left( \hat{c}_{k+Q}, \hat{c}_{k+2Q}, \ldots \hat{c}_{k+qQ} \right)
\]

\[
H_k = \begin{pmatrix}
\epsilon_{k+Q} & \Delta_Q & 0 & \ldots & 0 & \Delta_Q^* \\
\Delta_Q^* & \epsilon_{k+2Q} & \Delta_Q & 0 & \ldots & 0 \\
0 & \Delta_Q & \ldots & \ldots & \ldots & \ldots \\
\vdots & 0 & \vdots & \ldots & \ldots & \ldots \\
0 & \ldots & \ldots & \ldots & \ldots & \ldots \\
\Delta_Q^* & 0 & \ldots & 0 & \epsilon_{k+qQ} & \Delta_Q
\end{pmatrix}
\]

For the case |ΔQ|/t = 1 this is precisely the matrix form of Harper’s equation, applied by Hofstadter to the 2D electron gas in the presence of a magnetic field [8]. A plot of the eigenvalues against filling fraction has come to be known as Hofstadter’s butterfly (Figure 2). The eigenvalues of the matrix in Equation (2) constitute the possible energies of the system after the charge order has been established. In the presence of a charge modulation with period qa, the energies form q sub-bands, separated by energy gaps [23].

The contribution of each filled sub-band to the overall conductivity of the system is quantized in units of 2πe^2/h [4]. The quantization of conductance can be understood by realizing that the phase, θ, signifying the position of the charge modulation relative to the atomic lattice, is not fixed by the mean-field solution of Equation (2). The band structure can thus be drawn within a 2D space spanned by θ and k0, as indicated on the right of Figure 2 for the case p/q = 1/3. Each of these 2D sub-bands can be assigned a topological quantum number, the Chern number C1, by integrating the Berry curvature over this 2D space. In this specific problem C1 can also be found by solving a Diophantine equation [4, 25]. The quantized conductance is then the sum of the Chern numbers for all occupied sub-bands [4]. In the quantum Hall effect, the Hall conductivity σxy is given by (2πe^2/h)C1. In the present case, the response function associated with the topological invariant is instead a ‘quantized adiabatic particle transport’ [26, 27]: adiabatically cycling θ through a full period results in the transfer of an integer number C1 of electrons across the length of the 1D chain. Since θ describes the position of the charge modulation with respect to the atomic lattice, this cycling of θ corresponds to rigidly sliding the charge-ordered state through a single wavelength.

The Chern numbers for the commensurate charge-ordered patterns which make up the left of Figure 2 are indicated by its color scale. Only commensurate charge density modulations appear in the figure, but properties of the incommensurate charge-ordered states can be inferred from its large-scale structure. In particular, approaching an irrational value of the filling fraction as the limit of a series of rational fillings, it is clear that there must be large gaps (for example at the wings of the butterfly) even for incommensurate charge density modulations. We can then use the self-similarity of the fractal to see that any incommensurate state at irrational filling fraction η in Figure 2 must have the same nonzero sum of Chern numbers for its filled bands as the state with commensurate order at the rational filling p/q = η − ε, with ε → 0, which approaches η arbitrarily closely. For example, a close inspection of the underside of the butterfly’s lower left wing reveals a constant lining of C1 = 1 for all commensurate states filled up to the wing, which must therefore also apply to the incommensurate states interpolating between them.

The Chern numbers characterizing the quasicrystalline state can take on any integer value depending on the value of the (irrational) filling fraction. This observation does not contradict the usual topological classification, as the Berry curvature giving rise to the Chern numbers is integrated over both k and θ, so that the relevant parameter space is 2D. The quantized transport and the corresponding edge states are properties of the entire 2D family of locally isomorphic charge-ordered states related by translations over θ, in direct analogy to the case of the optical waveguide experiments [10, 16].

Experimental Realization—In the analysis of the spectrum of Equation (2) we imposed a fixed value of ΔQ(h), and did not take into account the self-consistency condition ΔQ(h) = 2h \sum_k \langle \hat{c}_{k+Q}^\dagger \hat{c}_k \rangle. Although the self-consistency requirement can affect the sizes of the differ-
FIG. 3. **Left:** the allowed energies (scale truncated at $[-2.5\,\hbar, 2.5\,\hbar]$) as a function of filling fraction $p/q \in (0, 1)$, $q \in [2, 18]$ for the self-consistent solution of Equation (1) with $\hbar = t$. Each sub-band is adiabatically connected to the corresponding sub-band in Figure 2. This implies that the sums of Chern numbers of filled sub-bands, indicated by the color scale, are the same. **Right:** the Berry curvatures (color coded and displaced for clarity) for the three sub-bands at 1/3 filling. Although the shapes are different to those in Figure 2, the integrals over them remain 1, −2, 1 in units of $2\pi$. The larger gaps that correspond for consecutive sub-bands will be closed when they become of the order of the effective energy scale set by local impurities, disorder, or lattice defects. When this happens, the Chern numbers of the two merging bands are unaltered by deformations of $|\Delta Q(h)|$. The self-consistent solution at a given interaction strength $\hbar$ therefore has the same topology as the standard Hofstadter case of Figure 2.

Real materials cannot maintain the infinite fine structure of a fractal band. Instead, the gaps separating consecutive sub-bands will be closed when they become of the order of the effective energy scale set by local impurities, disorder, or lattice defects. When this happens, the Chern numbers of the two merging bands are added together to yield the Chern number describing the combined sub-band [25]. The larger gaps that correspond for example to the wings of the butterfly may be expected to survive in clean enough systems at low temperatures. For the corresponding filling fractions, the sum of Chern numbers must then still be nonzero, even for incommensurate charge-ordered states in the presence of defects and disorder.

A second effect of the presence of impurities in real materials is their ability to pin the charge order in place, inhibiting the freedom of the modulated pattern to slide along the atomic lattice. It is well established, however, that an applied voltage may be used to overcome the pinning potential in incommensurate charge-ordered materials, allowing them to conduct even in the presence of pinning centers [23, 29, 31]. The current produced in such a way includes both the effects of adiabatic particle transport originating in the band topology, and the effects of non-adiabatic band-mixing. Since we are here only interested in the former effect, we propose an alternative way of accessing the quantized particle transport in both commensurate and incommensurate charge-ordered states, based on the use of atomic condensates in optical lattices [32]. Such systems are more readily controllable and defect-free than 1D chains in actual materials. Indeed, the Harper equation has recently been realized in a cold atom setup [28]. Impurities (including the confinement potential) will inevitably exist, but we propose to use them to our advantage. An impurity can be simulated by altering the potential on a single site, providing for example an attractive center which locks one of the maxima of the charge density modulation into place. Provided that this intentional pinning does not disrupt the large-scale gap structure, the topology of the sub-band remains unaffected. By manipulating the impurity location, the charge density modulation can be dragged along the optical lattice, continuously cycling the value of $\theta$ along its entire range. As the corresponding charge-ordered pattern moves through one wavelength, it should be possible to measure the transport of an integer number ($C_1$) of electrons across the length of the optical lattice.

**Conclusions** - In this letter we have shown the equivalence between families of 1D locally isomorphic quasicrystals and a mean-field model of incommensurate charge order in 1D materials. Both exhibit the same set of topological quantum numbers normally ascribed to 2D systems, in agreement with recent results on optical waveguide experiments [9, 13]. Using the self-similarity of the fractal pattern of allowed energies to interpret incommensurate charge-ordered materials as the limit of a series of commensurate cases, it becomes clear that these Chern numbers arise from an integral of the Berry curvature over a 2D space covering the entire family of locally isomorphic charge-ordered states. It is therefore the family of modulation patterns, rather than any individual quasicrystalline state, which is characterized by a 2D topological quantum number [10].

We find no conceptual difference between the topologies of the sub-bands for commensurate and incommensurate charge order, in the sense that the properties of an incommensurate charge-ordered state can be determined from the limit of a series of commensurate phases using the large-scale structure of the Hofstadter spectrum. The topological properties of the system survive in the full self-consistent solution of the model, and are robust against the inclusion of weak fluctuations, defects and impurities. The large-scale structure of the Hofstadter spectrum guarantees the possibility of quantized adiabatic particle transport in charge-ordered materials, as long as the gap size remains nonzero.

Finally, we propose an experimental test of the topological properties of charge-ordered materials by exploiting the possibility of directly controlling the phase of a
charge density modulation in atomic condensates in optical lattices. We predict that dragging the phase through a full period by this method will lead to the transfer of a quantized number of electrons across the system, providing a novel standard of conductance.

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[24] Our image is a slight variation on Hofstadter’s since we discard non-coprime fractions. An example is $p/q = 1/2$ which has 2 bands in our case but is a solid line in Hofstadter’s. Our color coding also implies a slight variation on statements made for example in Reference [28]: for $1/4$ filling the central bands touch, so their Chern numbers should be added. We note that in our case higher harmonics of the charge order would couple the relevant sub-bands and cause a splitting.
[32] We address the issue of quantized adiabatic particle transport in quasi-1D materials in an upcoming paper.