Spiral Fermi Surfaces in Quasicrystals and Twisted Bilayer Graphene: Signatures in Quantum Oscillations

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We show that electronic materials with quasiperiodic order can exhibit exotic “spiral Fermi surfaces” in two or three dimensions. These Fermi surfaces are self-intersecting, and characterized by a winding number of their surface tangent—a topological invariant—that is larger than one. Furthermore, we show that spiral Fermi surfaces can be probed directly through standard quantum oscillation experiments, arising from the semiclassical quantization of orbits. We compute the nature of the quantum oscillations in two experimentally relevant settings which give rise to spiral Fermi surfaces: a “nearly-free-electron” quasicrystal; and $30^\circ$ twisted bilayer graphene.

Quasiperiodic systems are long-range ordered and yet non-periodic [1–4]. This places them in a fascinating intermediate regime between periodic and disordered materials [5, 6]. The lack of periodicity rules out Bloch’s theorem, and can lead to localisation analogous to that induced by true disorder [7–10]. Yet their long-range order allows for the appearance of phenomena typically associated with periodicity, including Bloch oscillations [11–13] and quantum oscillations [14–16]. Furthermore, there has been a growing interest in their interplay with topology [17–23].

Although the quasiperiodic potential precludes the existence of a periodic Brillouin-zone (as in a conventional periodic lattice), aspects of band theory can emerge in the generally densely gapped spectrum [24–26] due to breakdown phenomena [13, 15, 27, 28]. This leads to an effective band structure defined in a pseudo-Brillouin-zone [29] which does not have a periodic structure. Within this construction, quantities related to band topology, such as the Berry curvature, can be defined [13]. However, most notably, the lack of periodicity of the pseudo-Brillouin-zone causes the bands themselves to have a complex self-intersecting structure [13]. This structure arises from a unique topology, in which the cyclic variation of a parameter leads to a change in the energy level, providing an example of a “spiral holonomy” [30].

In this paper we show that, in this effective description, quasicrystalline lattices generically lead to a novel form of Fermi surface, which we therefore call a “spiral Fermi surface”, in which the surface crosses itself one or more times. We show that spiral Fermi surfaces arise in two simple experimentally relevant models: a nearly-free-electron quasicrystal [31–34] and $30^\circ$ twisted bilayer graphene [35–37]. We calculate the quantized energy levels in an external magnetic field, via semiclassical quantization of the orbits on the spiral Fermi surfaces. We show that characteristic features of the spiral Fermi surfaces of these materials can be probed using standard quantum oscillation studies at experimentally accessible parameters.

Specifically, the topology of the spiral Fermi surfaces is classified using the turning number, $N_t$, which is defined as the winding number of the surface tangent—an invariant for two-dimensional plane curves [38, 39]. A Fermi surface that can be smoothly deformed to a circle has $N_t = \pm 1$ and is considered trivial, while all other turning numbers are considered nontrivial. Quantum oscillation parameters are determined by semiclassical quantisation [40–43],

$$\ell_B^2 S(E) = 2\pi (n + \gamma),$$  \hspace{1cm} (1)  

where $\ell_B = \sqrt{\hbar/eB}$ is the magnetic length, $S(E)$ is the area swept out by the wavepacket in reciprocal space and $2\pi\gamma$ is a phase offset. We highlight that the Fermi surface topology naturally appears in the Maslov contribution [43–47], $\varphi_M = \pi N_t$, to the phase offset $2\pi\gamma = \varphi_M - \varphi_B$, with $\varphi_B$ the Berry phase [43, 48]. This experimental technique is both well founded [42] and much used in the study of novel materials such as topological insulators [49–53] and semimetals [54, 55].

Nearly-free-electron quasicrystal. Our first model is for a nearly-free-electron quasicrystal with an axis of five-fold rotational symmetry. This is an approximation to icosahedral [1] and decagonal quasicrystals [56] that have nearly-free-electron qualities [24, 28, 34, 57–59], such as the various aluminium based quasicrystals studied in ARPES [31–33]. The model here amounts to keeping the 10 dominant Fourier components, (i.e. the brightest spots in the diffraction pattern), in particular those with Bragg planes (perpendicular bisectors) that intersect the free electron Fermi surface. We study the two-dimensional single particle spinless Hamiltonian,

$$\hat{H} = \vec{p}^2/2m + V(\hat{r}),$$  \hspace{1cm} (2)  

where $\vec{G}_j \equiv 2\kappa (\cos 2\pi j/5, \sin 2\pi j/5)$ and $V_0$ is the strength of each individual Fourier component, which is assumed to satisfy the nearly-free-electron limit, $V_0 \ll E_\kappa \equiv \hbar^2\kappa^2/2m$. This model was previously studied in...
The quantum oscillations in a nearly-free-electron quasicrystal (a) Sketch of our model for a nearly-free-electron quasicrystal, consisting of 10 Fourier components (largest blue points) at momenta ±G_i (red arrows) which have Bragg planes (grey dotted lines) forming a pseudo-Brillouin-zone (orange decagon) and which intersect the free electron Fermi surface (blue circle). All combinations of these 10—the reciprocal lattice of periodic systems—covers k-space densely (smaller blue points). (b) The semiclassical trajectories in an external magnetic field (solid blue curves) drift along the free electron Fermi surface (dotted circle) with scattering by each G_i at the pseudo-Brillouin-zone boundary. (c) These semiclassical trajectories are seen as constant energy contours (blue curves) of an ‘effective band structure’, which is shown for V_0 = 0.1E_κ. (d) The resulting spiral Fermi surface with nontrivial turning number of N_t = 2.

Ref. 13 and found to exhibit a spiral holonomy. We therefore only briefly review the required results, with a focus on how a topologically nontrivial Fermi surface arises.

The analysis of this model relies on the nearly-free-electron limit. This tells us that the free electron Fermi surface (the blue circle shown in Fig. 1a) remains mostly unchanged except for the opening of gaps proportional to V_0 at intersections with Bragg planes to ±G_i (dotted lines in Fig. 1a) and also gaps from combinations of n Bragg reflections. Since all combinations of G_i form a dense set in k-space (shown in Fig. 1a), the set of all associated gaps will also be dense. Crucially these gaps form a distinct hierarchy, Δ_{gap}^n ∝ (V_0/E_κ)^n. Thus, for V_0/E_κ small, one can choose a magnetic field that removes (n + 1)th order gaps via magnetic breakdown, while keeping nth order gaps. The probability of magnetic breakdown is given by [47, 60–63],

\[ P_{MB} = e^{-\frac{\pi a b \phi_B}{\hbar}}, \]

where a and b are the axes of the avoided crossing hyperbola. The simplest scenario—and that which will result in a spiral holonomy—is the regime of fields in which only first order gaps are kept,

\[ \left( \frac{V_0}{E_\kappa} \right)^4 \ll \frac{\hbar \omega_c}{E_\kappa} \ll \left( \frac{V_0}{E_\kappa} \right)^2, \]

where ω_c ≡ eB/m is the cyclotron frequency. We refer to this as the ‘first order regime’ of fields. The relevant gaps in this regime are along the pseudo-Brillouin-zone edges (yellow decagon in Fig. 1b).

Having specified an appropriate regime of magnetic fields—the first order regime—the semiclassical trajectories can be intuitively found by tracing a path along the unperturbed free electron Fermi surface and making jumps at intersections with relevant Bragg planes. This procedure is shown in Fig. 1b for a wavepacket that is initially localised at the top of the pseudo-Brillouin-zone in the free particle state |k⟩. This state proceeds clockwise around the free electron Fermi surface until it encounters the Bragg plane to G_1, at which point it is scattered into the state |k − G_1⟩. Continuing in this manner the wavepacket is scattered a total of five times between the following states

\[ |k⟩ \rightarrow |k − G_1⟩ \rightarrow |k − G_1 − G_3⟩ \rightarrow |k + G_2 + G_4⟩ \rightarrow |k + G_4⟩ \rightarrow |k⟩ \]

after which the wavepacket returns and can be quantised according to (1).

By projecting onto the above subset of states we find an effective band structure, as shown in Fig. 1c. The semiclassical trajectories described qualitatively above (blue curves in Fig. 1b) can now be seen quantitatively as the constant energy contours of this band structure shown Fig. 1c-d. The turning number can be computed by using a sum over the extremal points (points with vertical tangent), N_t = \frac{1}{2} \sum \nu_i, where \nu_i = ±1 for an extremal point with anticlockwise (clockwise) orientation. For the Fermi surface in Fig. 1d, there are four extremal points (yellow dots) with anticlockwise orientation. This gives a turning number of, N_t = 2, implying that this Fermi surface is nontrivial. This spiral Fermi surface (with N_t = 2) does not require fine tuning of the Fermi energy, unlike the “twisted Fermi surface” (with N_t = 0) of a tilted Weyl point [39].

We highlight two key signatures of this nontrivial turning number for quantum oscillations. The first is for the phase offset 2πγ in the semiclassical quantisation. Conventional Fermi surfaces can be deformed to a circle, which means \varphi_M = π and \gamma = 1/2, with deviations from this indicating a non-zero Berry phase. Here, N_t = 2, results in \varphi_M = 2π, which means \gamma = 0 for zero Berry phase. The second signature is related to the ‘magnetic breakdown transition’ between first order and second order field regimes, shown in Fig. 2. As this transition
occurs at a fixed Fermi energy and between an odd number of frequencies (γ) and an even number (α and β), at least one frequency must be associated with a Fermi surface with nontrivial turning number (we provide details for this argument in the Supplemental Material [64]).

In order to address these results experimentally, one must consider three key parameters: the Fermi energy $E_F$, the field $B$ and the potential $V_0$. In typical nearly-free-electron quasicrystals, $E_F$ is already at the required location—with the free-electron Fermi surface intersecting the pseudo-Brillouin-zone boundary [65]—therefore little to no doping should be required. For the model parameters used in Fig. 2, the flux density required to reach the first order regime is small compared to the electron density. Using typical experimental parameters of $\kappa = 1.3 \text{Å}^{-1}$ and $m = m_e$ (the free electron mass) [32], this occurs for fields of $B \approx 10 \text{T}$—a regime attainable experimentally. The required potential $V_0$, however, provides the most severe constraint experimentally. The calculation of the magnetic breakdown transition in Fig. 2 allows us to quantify the maximum allowed $V_0/E_\kappa$—for a ratio that is too large the two regimes (first and second order) are not distinguishable. Using this criteria we find a maximum of $V_0/E_\kappa \approx 0.02$, which corresponds to a gap at the pseudo-Brillouin-zone edge of approximately $0.2 \text{eV}$.

**Twisted Bilayer Graphene.**—Our second model is for 30°, twisted bilayer graphene, a system that has recently seen its first experimental realisation [35–37]. This incommensurate superstructure satisfies the typical definition of a quasicrystal [3] in that its diffraction pattern contains sharp peaks possessing an $n$-fold symmetry (here $n = 12$), and requires more basis vectors (four) than dimensions (two) in order to be indexed [66]. The quasiperiodic structure of the diffraction peaks is sufficient to cause the effective band structure to exhibit a spiral Fermi surface with a highly nontrivial turning number of $N_t = 5$.

To show this, we use the model of twisted bilayer graphene developed in Ref. 67. This takes a standard nearest neighbour tight-binding Hamiltonian $H_\parallel$ for each layer, which is off-diagonal in a Bloch basis, $|k, X\rangle$, with $X = A, B$ sublattice indices [68],

$$
(k, A)|H_\parallel|k, B\rangle = -t \sum_{i=1}^{3} e^{-i k \cdot \rho_i},
$$

where the vectors $\rho_i$ connecting nearest neighbours in layer 1 are rotated by 30° with respect to those in layer 2. Tunnelling between the layers causes a Bloch state from layer 1 with crystal momentum $k$ to be coupled to all those from layer 2 with crystal momentum $\hat{k} = k + G - \hat{G}$ [67],

$$
\langle k, \hat{k} | H_\perp |k, X\rangle = -t_\perp (k + G) e^{-i G \cdot \tau_X + i \hat{G} \cdot \tau_{\hat{X}}} ,
$$

where a tilde (no tilde) denotes layer 2 (1), $G$ is a reciprocal lattice vector, $\tau_X$ are position vectors of the sublattice sites within the unit cell, and $t_\perp (k)$ is radially symmetric and decays exponentially for $k$ beyond the first Brillouin-zone [69].

We analyse this model by assuming a weak coupling between the two layers, $t_\perp (k) \ll t$. As with the nearly-free-electron assumption in the previous section, this assumption is key to deriving meaningful semiclassical trajectories. In particular this allows us to assert that the Fermi surfaces of each layer will be little affected, except at degenerate points that satisfy,

$$
E(k) = \tilde{E}(\hat{k}), \quad k + G = \hat{k} + \hat{G},
$$

where $E(k)$ and $\tilde{E}(\hat{k})$ are the bandstructures of the unperturbed layers 1 and 2. This is considered a first order coupling, as a gap will open proportional to $t_\perp$. However there will also be gaps opened due to second order processes that couple a layer to itself at the following

![FIG. 2. Quantum oscillation frequency spectrum across the transition between second order and first order field regimes.](a) Density plot of the frequency spectrum (with frequencies given as a ratio of the spiral holonomy frequency $E_\gamma$) as a function of magnetic field (given in terms of $n_0 = eB/h$ and $n_{2D} = k_F^2/2\pi$), for the parameters $V_0/E_\kappa = 0.01$ and $E_F/E_\kappa = 1.01$. A transition is seen from a single frequency (γ) at high fields ($n_0/n_{2D} \approx 10^{-4}$) to a pair of frequencies (α and β) at lower fields ($n_0/n_{2D} \approx 10^{-7}$). (b) Plot of the magnetic breakdown probabilities at second order gaps (orange curve) and first order gaps (blue curve). (c) A selection of semiclassical contours used to label frequencies. For intermediate fields a complex mix of frequencies are present that can be labelled using intermediate contours such as $\epsilon_1$ and $\epsilon_2$.}
FIG. 3. Quantum oscillations in incommensurate 30° twisted bilayer graphene. (a) Sketch of each layer’s Brillouin-zone, blue is referred to as layer 1 and red as layer 2. (b) The Fermi surface of layer 1 is coupled to the Fermi surface of layer 2, in addition to all possible translations of this Fermi surface by reciprocal lattice vectors from layer 1. (c) For sufficiently large doping, the Fermi surface of layer 1 intersects that of layer 2, allowing a semiclassical trajectory that jumps from layer 1 to layer 2. This repeats a total of 12 times before returning to be quantised by semiclassical quantisation. (d) The Fermi surface of an effective model, shown for the experimental parameters given in Ref. 70.

with these gaps proportional to $t_\perp^2/t$. For simplicity we choose to work in a field regime in which second order, intra-layer gaps can be ignored, while inter-layer are kept—which can be safely assumed to exist given the weak coupling assumption. However, this still leaves a dense set of gaps given by (8). Fortunately, many of these are exponentially small due to the $k$ dependence of $t_\perp(k)$, as shown in Fig. 3b. We therefore choose a field such that those gaps opened by the exponential tail of $t_\perp(k)$ beyond the first Brillouin-zone are ignored. In this regime the doping necessary to see interlayer effects is then simple to see as that at which the original Fermi surfaces of the two layers intersect.

Having determined a suitable field regime and doping, we derive the semiclassical trajectories by simply tracing a path along the unperturbed Fermi surfaces and switching between layers at the relevant intersections. If one begins this process, as shown in Fig. 3c, on layer 1 (blue contour), the wavepacket will progress anticlockwise before jumping to layer 2 (red contour). Here it essentially repeats this contour again, now rotated by $5\pi/6$. This occurs a total of 12 times, resulting in a trajectory that winds the centre a total of 5 times, as shown in Fig. 3c. This is reflected in the effective Fermi surface shown in Fig. 3d, shown for typical model parameters. The turning number is computed by summing over the extremal points (as discussed in the previous section). For each $2\pi$ winding about the centre there are two extremal points with anticlockwise orientation. As the Fermi surface winds the centre 5 times, there are a total of 10 extremal points with anticlockwise orientation, which means $N_t = 5$, and therefore this Fermi surface is nontrivial.

Since the turning number in this case is odd, the two signatures highlighted in the previous section for a nearly-free-electron quasicrystal do not apply here—$\gamma = 1/2$, which is indistinguishable from the trivial case and breakdown transitions cannot identify odd turning numbers [64]. Instead, the key signature here is in the dependence of the quantum oscillation frequency on doping, as shown in Fig. 4. Below a critical doping of approximately 2 eV, the Fermi surfaces of each layer do not

FIG. 4. Phenomenology for 30° twisted bilayer graphene. Plot of quantum oscillation frequencies as a function of doping away from charge neutrality, using the same parameters given in Fig. 3. (Inset) Each frequency is identified with a different starting point on the unperturbed ‘Dirac’ Fermi surface, the purple contours are those with nontrivial turning number and have a larger than naively expected frequency.
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[70] We use the following parameters for bilayer graphene, $t = -2.7eV$, $V_{\text{pp}}^0 = 0.48eV$, $a = 0.246nm$, $d = 0.335nm$, $r_0 = 0.184a$ and $\kappa = 2\pi/\sqrt{3}a$, from Ref. 67.

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Identifying Nontrivial Turning Numbers from Magnetic Breakdown

Here we derive a general statement concerning the relationship between magnetic breakdown transitions and the presence of nontrivial turning numbers. That is: given a magnetic breakdown transition that occurs at fixed Fermi energy, the sum of all turning numbers must be conserved, which implies that at least one Fermi surface contour has nontrivial turning number (|N| ≠ 1) if there is a change in the number of frequencies modulo 2.

We first highlight that magnetic breakdown occurs in two varieties, referred to as interband and intraband. As their names suggest, interband occurs between two bands, typically with an avoided crossing, whereas intraband occurs in a single band, typically at a saddle point. Both types can be described by a two-in-two-out breakdown vertex, as shown in Fig. S1, in which case they simply label the two unique orientations of incoming and outgoing vertices (up to rotations).

The associated scattering matrices that connect incoming and outgoing edges are as follows. For interband one has (using the notation in Fig. S1),

\[
\begin{pmatrix} c_1 \\ c_2 \end{pmatrix} = S_{\text{interband}} \begin{pmatrix} \rho c_1^+ \\ -\rho c_2^+ \end{pmatrix}
\]

with scattering matrix [73],

\[
S_{\text{interband}} = \begin{pmatrix} \tau \rho^{i(\omega+\theta)} & -\tau e^{-i(\omega+\theta)} \\ \rho & -\tau e^{-i(\omega+\theta)} \end{pmatrix},
\]

where \( \rho = e^{-\tau \mu}, \tau \equiv \sqrt{1-\rho^2}, \omega = \mu - \mu \ln \mu + \text{arg} \Gamma(i\mu) + \pi/4, \theta \) is the phase of the matrix element that opened the avoided crossing, and \( \mu = \frac{1}{2} ab \ell_B^2 \) with \( a \) and \( b \) the hyperbolic axes of the avoided crossing. Whereas for intraband one has,

\[
\begin{pmatrix} c_\text{intra} \\ c_\text{inter} \end{pmatrix} = S_{\text{intra}} \begin{pmatrix} c_\text{inter}^+ \\ -c_\text{intra}^+ \end{pmatrix}
\]

with scattering matrix,

\[
S_{\text{intra}} = \begin{pmatrix} \mathcal{R} & \mathcal{T} \\ \mathcal{T} & \mathcal{R} \end{pmatrix}
\]

where,

\[
\mathcal{T}(\mu') = e^{i\phi(\mu')} \frac{e^{\pi \mu'/2}}{\sqrt{2 \cosh(\pi \mu')}}
\]

with \( \mathcal{R}(\mu') = -ie^{\pi \mu'} T(\mu'), \phi(\mu') = \text{arg} \{\Gamma(i\mu')\} + \mu' \log |\mu'| - \mu', \) and \( \mu' = \text{sgn}(E) \frac{1}{2} ab \ell_B^2, \) with \( ab \) as for interband and \( E \) is the energy relative to the crossing point of the saddle point.

A change in the Fermi surface connectivity occurs when the scattering matrix transitions between diagonal and off-diagonal. Crucially, this can occur at fixed Fermi energy for interband. Whereas for intraband, the Fermi energy is required to cross the saddle point. For interband one has,

\[
\mu \rightarrow \infty, \quad S_{\text{interband}} \rightarrow \begin{pmatrix} e^{i\theta} & 0 \\ 0 & -e^{i\theta} \end{pmatrix}
\]

\[
\mu \rightarrow 0, \quad S_{\text{interband}} \rightarrow \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}
\]

whereas for intraband one has,

\[
\mu' \rightarrow \infty, \quad S_{\text{intra}} \rightarrow \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}
\]

\[
\mu' \rightarrow 0, \quad S_{\text{intra}} \rightarrow \begin{pmatrix} 1/\sqrt{2} & -i/\sqrt{2} \\ -i/\sqrt{2} & 1/\sqrt{2} \end{pmatrix}
\]
\[
\mu' \to \infty, \quad S_{\text{intraband}} \to \begin{pmatrix} 0 & -i \\ -i & 0 \end{pmatrix}.
\] (19)

One can therefore distinguish the type of magnetic breakdown by assessing whether or not magnetic breakdown occurs at fixed Fermi energy.

Furthermore, the sum of all turning numbers is conserved across an interband transition. Denoting the, \( n \), turning numbers before the transition, \( N_i^t \), and the, \( n' \), turning numbers after by, \( N_i'^t \), one has,

\[
\sum_{i=1}^{n} N_i^t = \sum_{i=1}^{n'} N_i'^t \mod 2 \tag{20}
\]

An example is shown in Fig. S1, in which a figure of eight curve with a nontrivial turning number of zero is split into two trivial curves with turning numbers, \( \pm 1 \), as \( \mu : 0 \to \infty \). For intraband transitions, the turning number is not conserved, with an example shown in Fig. S1.

Given that the sum of all turning numbers is conserved, a statement about the presence of nontrivial turning numbers can be made using the following argument. First, we assume all turning numbers are trivial before and after a transition, \( |N_i^t| = |N_i'^t| = 1 \) for all \( i \). Then we have,

\[
\sum_{i=1}^{n} N_i^t = n \mod 2,
\] (21)

and,

\[
\sum_{i=1}^{n} N_i'^t = n' \mod 2.
\] (22)

Since the sum of all turning numbers is conserved (20),

\[
n = n' \mod 2.
\] (23)

However this is not necessarily true, as in the case shown in Fig. S1. We therefore have a contradiction: if, \( n \neq n' \mod 2 \), the assumption that all turning numbers before and after are trivial must be incorrect. Therefore at least one turning number must be nontrivial.

More precisely, this test identifies that there is at least one even turning number (nontrivial by definition). However, a nontrivial odd turning number may be present and would not be identified by this method. Therefore a change in parity of the total number of turning numbers is a necessary but not sufficient condition for the presence of a nontrivial turning number.

To summarize, if magnetic breakdown occurs at fixed Fermi energy, this must be due to an interband transition. In this case, the sum of all turning numbers is conserved. Furthermore, if the total number of turning numbers (or frequencies in the quantum oscillations) changes modulo 2, there must be at least one nontrivial turning number.

\[
\mu' \to \infty, \quad S_{\text{intraband}} \to \begin{pmatrix} 0 & -i \\ -i & 0 \end{pmatrix}.
\] (19)

One can therefore distinguish the type of magnetic breakdown by assessing whether or not magnetic breakdown occurs at fixed Fermi energy.

FIG. S2. Scattering matrix generalisation of semiclassical quantisation. Sketch showing how a generalised semiclassical quantisation is applied to the spiral holonomy contours. Here the wavepacket undergoes partial magnetic breakdown at avoided crossings (pink disks) described by scattering matrices, \( S_i \), but follows classical trajectories along intermediate contours with phase evolution described by diagonal matrices, \( \Lambda_i \).

**Numerical Approach for Magnetic Breakdown Spectrum**

In order to compute the quantum oscillation frequency spectrum in Fig. 2, we use a generalisation of semiclassical quantisation [47, 72]. In which we replace the self intersections (in the first order regime) of the Fermi surface contours with interband scattering matrices given in (11), and as shown in Fig. S2. A diagonal matrix accounts for phase evolution along intervening contours,

\[
\Lambda = \begin{pmatrix} e^{i\Omega_1} & 0 \\ 0 & e^{i\Omega_2} \end{pmatrix},
\] (24)

where \( \Omega_i \equiv \ell_B^2 S_i + \varphi_{M_1} + \varphi_{B_1} \), is the sum of area, Maslov and Berry contributions. While the total unitary evolution is given by the product

\[
U = \prod_{j=1}^{5} \Lambda_j S_j.
\] (25)

By requiring single-valuedness, \( U\psi = \psi \), an equation that generalises semiclassical quantisation for intermediate breakdown is provided by,

\[
\det(U - I) = 0.
\] (26)

This equation recovers the usual semiclassical quantisation (1) in both limits \( P_{\text{MB}} \to 0, 1 \).

The spectrum is then computed by constructing the density of states at the Fermi energy as a function of \( 1/B \). Here this consists of delta peaks centred at each solution in \( B \) to (26), as shown in Fig. S3a. The discrete Fourier transform [74] is then taken over a finite range of \( 1/B \) (a small non-zero broadening is used for the delta peaks.
FIG. S3. Examples of frequency spectrum for three regimes of magnetic field. (a) Fourier transform of density of states at fixed Fermi energy (shown inset). These a slices of the density plot in Fig. 2 at the field values (top to bottom): \( n_\phi/n_{2D} = 10^{-4} \), \( n_\phi/n_{2D} = 10^{-6} \) and \( n_\phi/n_{2D} = 10^{-7} \). (b) Semiclassical contours used to label frequency spectrum plots in (a).