Hofstadter Butterfly and Integer Quantum Hall Effect in Three Dimensions

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For a three-dimensional (3D) lattice in magnetic fields we have shown that the hopping along the third direction, which normally smears out the Landau quantization gaps, can rather give rise to a Hofstadter's butterfly *specific to 3D* when a criterion is fulfilled by anisotropic (quasi-one-dimensional) systems. In 3D the angle of the magnetic field plays the role of the field intensity in 2D, so that the butterfly can occur in much smaller fields. We have also calculated the Hall conductivity in terms of the topological invariant in the Kohmoto-Halperin-Wu formula, and each of σ_{xy}, σ_{zx} is found to be quantized.

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Among the effects of magnetic fields on electronic states, one of the most bizarre is Hofstadter's butterfly [1]. Namely, when a two-dimensional (2D) periodic system is put in a magnetic field, gaps not only appears between the Landau levels, but a series of gaps appears in a self-similar, fractal fashion against the magnetic flux ϕ, penetrating a unit cell in units of the flux quantum $\phi_0 = h/e$. From its derivation the butterfly is conceived to be specific to 2D.

Here we raise a question: can we have something like Hofstadter's butterfly in spite of, or even because of, a three-dimensionality (3D)? This may at first seem quite unlikely since a motion along the third direction (z) should wash out the butterfly gaps as well as Landau level gaps. Several authors have extended Hofstadter's problem to 3D in the last decade [2,3], and subbands are indeed shown to overlap or touch with each other.

Here we have found that an analog of Hofstadter's butterfly does indeed exist, which is, intriguingly, not a remnant of a 2D butterfly but *specific to* 3D, appearing under a certain condition that is fulfilled by anisotropic (quasi-1D) systems. The problem is solved by formally mapping the 3D Schrödinger's equation to 2D. The mapping indicates that the ratio of the magnetic fluxes penetrating two facets of the 3D unit cell plays the role of the magnetic flux in 2D, so that the 3D butterfly appears on the energy versus *tilting angle* of the magnetic field.

Once we have a butterfly, we can immediately ask ourselves how the *integer quantum Hall effect* should look on the butterfly. If one examines a theoretical reasoning from which the quantization in the Hall conductivity is deduced in the usual quantum Hall system, an essence is the presence of a gap in the energy spectrum. This was already indicated in a gauge argument by Laughlin [4] and elaborated by Thouless *et al.* [5] for periodic systems. There the quantized Hall conductivity for the Fermi energy E_F lying in a butterfly gap is identified to be a topological invariant.

For 3D Kohmoto, Halperin, and Wu have shown, following the line of the 2D work, that *if* there is an energy gap in a 3D system, then an integer quantum Hall effect should result when E_F lies in a gap [6,7]. Montambaux and Kohmoto have calculated the Hall conductivity in a case where a third-direction hopping opens some gaps [8].

Since the 3D butterfly found here has a recursive structure we question the systematics of the quantum Hall effect. The 2D-3D mapping evoked to derive the 3D butterfly indeed enables us to calculate the Hall topological invariants for the 3D butterfly. We have found that *each of* σ_{xy}, σ_{zx} *is quantized* in 3D.

Our model is one of noninteracting tight-binding electrons in a uniform magnetic field $\boldsymbol{B}$ described by the Hamiltonian

$$\mathcal{H} = \sum_{\langle i,j \rangle} (t_{ij} e^{i\theta_{ij}} c_i^\dagger c_j + \text{H.c.}), \tag{1}$$

in standard notations, where the summation is over nearest-neighbor sites with $t_{ij} = t_x, t_y, t_z$ along x, y, z, respectively, and $\theta_{ij} = (e/\hbar) \int_i^j \boldsymbol{A} \cdot d\boldsymbol{l}$ is the Peierls phase. Let us first recapitulate the 2D butterfly. In 2D with the Landau gauge $\boldsymbol{A} = (0, Bx)$, y is a cyclic coordinate, so that the wave function becomes $\psi_{lm} = e^{i\nu_y m} F_l$, where (l, m) labels (x, y) coordinates, and ν_y is the Bloch wave number along y. The Schrödinger equation then takes a form of Harper's equation,

$$-t_x(F_{l-1} + F_{l+1}) - 2t_y \cos(2\pi\phi l + \nu_y) F_l = EF_l, \tag{2}$$

where $\phi = Bab/\phi_0$ is the number of flux quanta penetrating a unit cell $= a \times b$. The energy spectrum becomes a butterfly for the ordinary isotropic case, $t_x = t_y$, while we note for later reference that the gaps are rapidly smeared out as the anisotropy is introduced, $t_y/t_x \to 0$, because the cosine potential weakens. Since t_x and t_y enter on an equal footing, the 2D butterfly appears when $t_x \approx t_y$.

Harper's equation in 3D can be derived in a similar way. For simplicity we consider a simple cubic lattice in a magnetic field $\boldsymbol{B} = (0, B\sin\theta, B\cos\theta)$ assumed to be on the yz plane [9]. The vector potential is then $\boldsymbol{A} = (0, Bx\cos\theta, -Bx\sin\theta)$, so that y, z are cyclic coordinates and the wave function becomes $\psi_{lmn} = e^{i\nu_y m + i\nu_z n} F_l$, where (l, m, n) labels (x, y, z). The Schrödinger equation is

$$-t_x(F_{l-1} + F_{l+1}) - [2t_y \cos(2\pi\phi_z l + \nu_y) + 2t_z \cos(-2\pi\phi_y l + \nu_z)]F_l = EF_l\,, \tag{3}$$

where two periodic potentials are superposed. Here $\phi_y(\phi_z)$ is the number of flux quanta penetrating the side of a unit cell ($= a \times b \times c$) normal to $y(z)$ [inset of Fig. 3(a)].

Although the spectrum in 3D does not usually have many gaps (aside from the trivial Bragg-reflection gaps), we find that a butterflylike structure does emerge for certain choices of (t_x, t_y, t_z), as typically displayed in Fig. 1(a) for $(t_x, t_y, t_z) = (1, 0.1, 0.1)$, a quasi-1D system. The spectrum is plotted against the angle θ of a magnetic field $(\phi_y, \phi_z) = 0.2(\sin\theta, \cos\theta)$ with $b = c$ assumed here. A structure akin to the 2D butterfly is seen in the bottom (or at the top) of the whole spectrum.

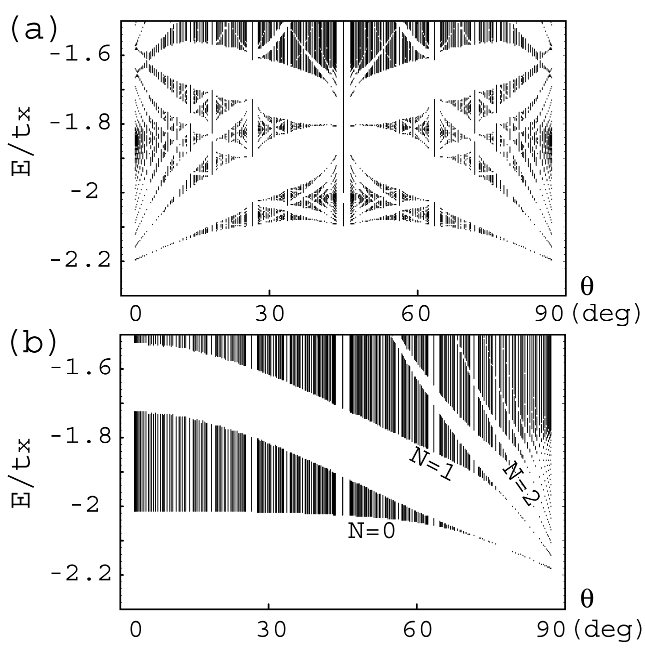

FIG. 1. The energy spectrum of a 3D system with $t_x{:}t_y{:}t_z = 1{:}0.1{:}0.1$ (a) or a 2D system with $t_x{:}t_y{:}t_z = 1{:}0.1{:}0$ (b), plotted against the angle θ in a magnetic field $(\phi_y, \phi_z) = 0.2(\sin\theta, \cos\theta)$.

One might consider this as a 2D butterfly surviving the third-direction hopping, but this is wrong as is evident from Fig. 1(b), where we turn off t_z to find that the spectrum coalesces to a series of broadened Landau levels. So we are talking about the butterfly *specific* to 3D rather than a remnant of a 2D counterpart.

We first explore the mechanism why the butterfly appears in 3D. For the periodic potentials in the 3D Harper equation, $V^{(1)}(l) \propto t_y \cos(2\pi\phi_z l + \nu_y)$ and $V^{(2)}(l) \propto t_z \cos(-2\pi\phi_y l + \nu_z)$, we assume that their periods are much greater than the lattice constant ($\phi_z, \phi_y \ll 1$). We also assume that $t_y\phi_z \gg t_z\phi_y$, i.e., local peaks and dips of the total potential $V^{(1)} + V^{(2)}$ are primarily those of $V^{(1)}$.

Then the potential wells of $V^{(1)}$, with a spacing $1/\phi_z$, feel the slowly varying $V^{(2)}$, and, since each well contains many original sites due to the assumption, we can talk about bound states of the well in the effective-mass sense. If wells are deep enough, several bound states appear per well and each state forms a tight-binding band (i.e., Landau band), and the equation (3) reduces to

$$-t'(J_{l'-1} + J_{l'+1}) - 2t_z \cos[-2\pi(\phi_y/\phi_z)l' + (\phi_y/\phi_z)\nu_y + \nu_z]J_{l'} = EJ_{l'}\,. \tag{4}$$

Here t' is the transfer integral between neighboring bound states, l' labels the well, $J_{l'}$ the wave function, and the cosine term represents $V^{(2)}$ at each minimum of $V^{(1)}$. The reduced equation has exactly the same form as that in 2D, Eq. (2), if we translate

$$\text{3D:}(t_x, t_y, t_z, \phi_y, \phi_z) \to \text{2D:}(t', t_z, \phi_y/\phi_z)\,. \tag{5}$$

Since the butterfly is a hallmark of an isotropic 2D case, one can predict that the spectrum in 3D should exhibit a butterfly when $t' \approx t_z$. So a finite t_z is *indispensable,* and the butterfly is in fact washed out for $t_z = 0$.

We can estimate t' (when there is $V^{(1)}$ alone) by converting Harper's equation to a differential equation for a continuous variable $\tilde{l} \equiv 2\pi\phi_z l$ in the effective-mass sense, which turns out to contain a combination t_y/ϕ_z^2 only (with $t_x = 1$, a unit of energy). Since t' is a matrix element of $V^{(1)} \propto t_y$, we have a simple scaling law,

$$t' = 2t_y f(t_y/\phi_z^2)\,. \tag{6}$$

The value of t' differs from one Landau band to another, where middle bands, with weaker binding, have larger t'.

We have numerically calculated $t'(t_y, \phi_z)$ for the lowest band. This, combined with the scaling, is shown in Fig. 2. If we plug in the condition for the butterfly, $t_z \simeq t'$, the plot may be regarded as indicating how to adjust ϕ_z to have a butterfly for given (t_y, t_z). We can see that the butterfly is

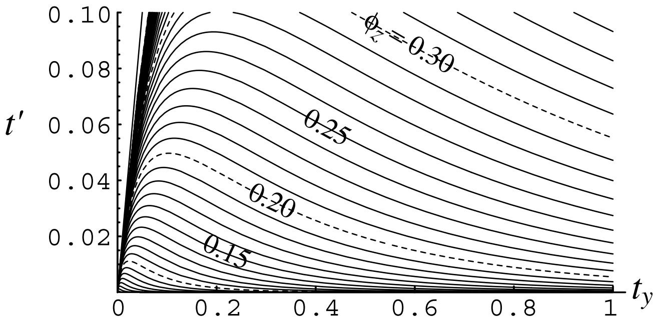

FIG. 2. The effective transfer $t'(t_y, \phi_z)$ for the lowest Landau band in units where $t_x = 1$. By plugging $t' \simeq t_z$, the plot serves to identify appropriate values of ϕ_z to realize the butterfly for given (t_y, t_z).

restricted to the case with $t_y, t_z \ll 1(= t_x)$, i.e., quasi-1D systems [10]. In other words, we cannot satisfy $\phi_z \ll 1$ when $t_y, t_z \approx 1$.

The plot also shows that the example above, $(t_x, t_y, t_z) = (1, 0.1, 0.1)$, $\phi_z \approx 0.2$, is indeed a right choice with $t' = 0.05 \approx t_z(= 0.1)$. The butterfly is symmetric in this example, which is an accident for $t_y = t_z$: in Harper's equation $V^{(1)}$ and $V^{(2)}$ exchange roles at $\theta = 45°$ for $t_y = t_z$. Around $t_y\phi_z \approx t_z\phi_y$, the argument in terms of the wells breaks down but a clear structure remains. This is because, although $V^{(1)} + V^{(2)}$ then exhibits a beat so that the barrier height separating the wells (hence t') varies from place to place, a change in the barrier height changes t' only slightly, since t' has a broad peak against t_y as seen in Fig. 2.

Would the 3D butterfly be experimentally realizable [11]? In principle, Fig. 2 shows that there exists appropriate (t_y, t_z) no matter how small ϕ_z may be, while the 2D butterfly requires $\phi \sim O(1)$ [12]. So, for a given lattice constant, the butterfly is more easily realized in 3D. In practice, (t_y, t_z) become smaller as ϕ_z decreases, and the energy scale [width of the Landau band $4(t' + t_z)$ with $t' \approx t_z$] shrinks when $\phi_z \to 0$, so that it will become harder to resolve the butterfly structure. For typical quasi-1D organic conductors such as $(TMTSF)_2X$ we have $t_x{:}t_y{:}t_z \sim 1{:}0.1{:}0.01$ with $a, b, c \sim 10$ Å, and we can estimate the required $\phi_z \sim 0.1$, an order of magnitude smaller than $\phi_z \sim 1$, and the energy scale $4(t' + t_z) \sim 20$ meV. $\phi_z = 0.1$ corresponds to $B \sim 400$ T, which is large but around the border of experimental feasibility [13].

To be more precise there are further restrictions on (t_y, t_z) to have butterflies. Binding of a well must be so strong that the transfer to second neighbors is negligible, which is shown to require $\sqrt{t_y} > \phi_z$. Also, different Landau bands should not be mixed, which requires $t_z < \phi_z\sqrt{t_y}$. Although all these conditions can be interpreted in the semiclassical quantization involving the cross sections of equipotential surfaces, the essential factor is, as seen, the hopping t' between adjacent cross-sectional orbits, which is outside the scope of the semiclassical quantization.

Now we come to our final goal of calculating the Hall conductivity for the 3D butterfly. The mapping used to derive the 3D butterfly enables us to accomplish this if we identify the topological invariants. In the Kohmoto-Halperin-Wu formula the Hall conductivity tensor is expressed as

$$\sigma_{ij} = -\frac{e^2}{2\pi h} \sum_k \epsilon_{ijk} G_k \tag{7}$$

when E_F is in a gap. Here ϵ_{ijk} is the unit antisymmetric tensor, $\boldsymbol{G} = \mu_1 \boldsymbol{a}^* + \mu_2 \boldsymbol{b}^* + \mu_3 \boldsymbol{c}^*$ with $\boldsymbol{a}^*, \boldsymbol{b}^*, \boldsymbol{c}^*$ being the primitive reciprocal lattice vectors, and μ_1, μ_2, μ_3 are topological invariants specifying each gap. For an orthogonal lattice we have simply $\sigma_{yz} = -\frac{e^2}{h}\frac{\mu_1}{a}$, $\sigma_{zx} = -\frac{e^2}{h}\frac{\mu_2}{b}$, and $\sigma_{xy} = -\frac{e^2}{h}\frac{\mu_3}{c}$.

The invariant integers are subject to a Diophantine equation,

$$\frac{r}{Q} = \lambda + \frac{P}{Q} n_x \mu_1 + \frac{P}{Q} n_y \mu_2 + \frac{P}{Q} n_z \mu_3 , \tag{8}$$

where we have assumed a rational magnetic flux, $(\phi_x, \phi_y, \phi_z) = \frac{P}{Q}(n_x, n_y, n_z)$ (P, Q: integers, n_x, n_y, n_z have no common divisors), with r the number of occupied bands and λ another topological invariant. Although the solution of the equation is not unique, Thouless *et al.* [5] argued for 2D that there is a restriction on the integers that makes the solution unique. In analogy with this Kohmoto *et al.* [7] conjecture the uniqueness of the solution in 3D, where the restriction reads $|\mu_1 n_x + \mu_2 n_y + \mu_3 n_z| < Q/2$.

We can then calculate the Hall conductivity for the 3D butterfly. We assume $\phi_x = 0$ and $t_y\phi_z \gg t_z\phi_y$, for which the effective flux in Eq. (4) is $\phi = \phi_y/\phi_z = n_y/n_z$, so each Landau band should split into n_z butterfly subbands. Consider E_F lying just above the mth subband in the lth Landau band from the bottom [i.e., $(ln_z + m)$ subbands altogether]. Each subband is shown to comprise P bands, so that the gap has an index $r = (ln_z + m)P$. When we substitute this in the Diophantine equation (8), we note that, since P and Q have no common divisors, λ must be a multiple of P; with the above restriction one has $\lambda = 0$, and we end up with $ln_z + m = n_y\mu_2 + n_z\mu_3$ for the 3D butterfly in the lower half of the entire band. From this we can determine μ_2, μ_3 for every gap in the 3D butterfly, as explicitly displayed in Fig. 3(a).

If we compare this with a corresponding plot for 2D in Fig. 3(b), we recognize a consequence of the 2D-3D mapping as a beautiful one-to-one correspondence between the Hall conductivities on 2D and 3D butterflies *as a whole* (i.e., for a set of topological invariants attached to the recursive gaps). Namely, the Hall conductivity in 2D [5] is given by $\sigma_{2D} = -\frac{e^2}{h}t$, where t is an integer in a 2D Diophantine equation $r = qs + pt$. If we compare this with the 3D Diophantine equation, the mapping dictates a correspondence $n_y \leftrightarrow p, n_z \leftrightarrow q, m \leftrightarrow r$, so that the invariant integers should translate (inset of Fig. 3) as

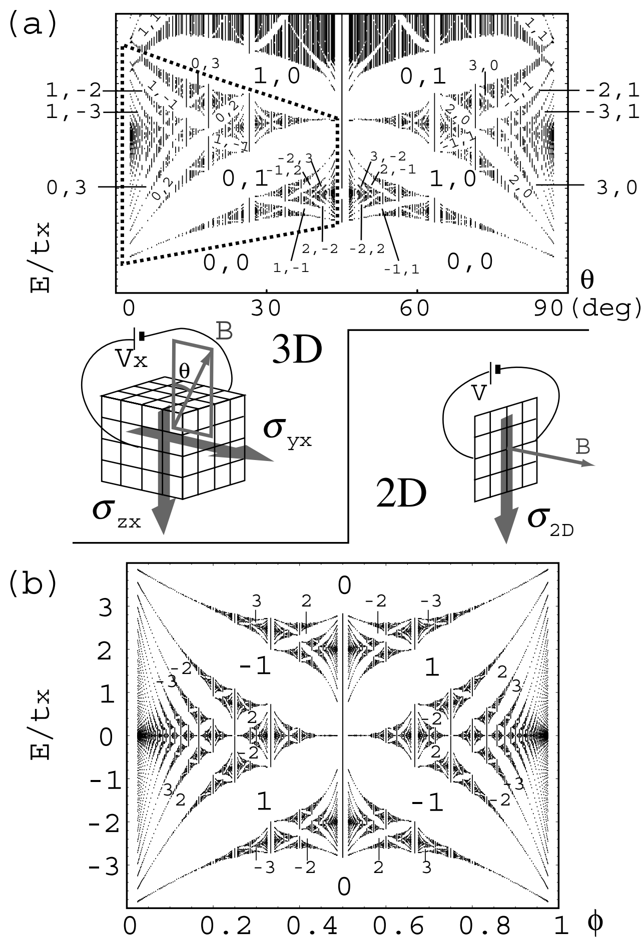

FIG. 3. (a) The conductivity $(\sigma_{xy}, \sigma_{zx}) = -e^2/h(\mu_3, \mu_2)$ is plotted on a 3D butterfly, where we display the topological invariants (μ_3, μ_2) for each gap. (b) The corresponding plot for $\sigma_{2D} = -(e^2/h)t$ on the 2D butterfly. The area enclosed by a dashed line in (a) corresponds to the 2D butterfly. μ_2 in (a) corresponds to t in (b), while μ_3 in (a) to s in Eq. (9).

$$\sigma_{xy} - l \longleftrightarrow s, \qquad \sigma_{zx} \longleftrightarrow \sigma_{2D}. \tag{9}$$

The discussions so far are for clean systems. We know that localization of electrons is necessary for the quantum Hall plateaus. In 2D, almost all the states are localized so that the Hall conductivity as a function of the electron concentration becomes step functions in the thermodynamic limit at $T = 0$. In 3D, there should be mobility edges, and plateaus will be formed between mobility edges. It is an interesting problem to study how the mobility edges appear in the 3D butterfly, which would require an entirely separate work. Even in 2D the step-function plateaus are smeared for a finite system size or a finite inelastic scattering length. For the usual 2D butterfly, a numerical study for a finite system [14] shows that we still have a nonmonotonic behavior as a sign for the butterfly when the disorder is not too strong, so we expect a similar behavior in the disordered 3D butterfly as well.

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[11] Magnetotransports such as the angle-dependent magnetoresistance oscillation have been experimentally observed in moderate magnetic fields (see, e.g., T. Ishiguro, K. Yamaji, and G. Saito, *Organic Superconductors* (Springer, New York, 1998), 2nd ed.
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