Hofstadter butterflies for flat bands

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Hofstadter's diagram (the energy spectrum against the magnetic field in tight-binding systems) is obtained for the models having flat one-electron band(s) that have originally been proposed for itinerant spin ferromagnetism. Magnetic fields preserve those flat bands that arise from a topological reason, while dispersions emerge in a singular manner for the flat bands arising from interference. This implies an anomalous orbital magnetism. [S0163-1829(96)51548-1]

Hofstadter's butterfly, or the Landau-quantized energy spectrum against the magnetic field in periodic systems, provides an intriguing example of the fractal spectrum in condensed matter physics. The quantum Hall effect for lattice fermions has also been discussed for these spectra.[1] Physically, the message is that when the magnetic field penetrating the unit cell of a two-dimensional lattice is q/p in units of flux quantum, we have essentially a p-band system. Accordingly the scaling of the integer quantum Hall effect, for instance, exhibits a peculiar structure for $p\neq 1$.[2]

Although the situation might seem essentially the same for complex lattices with the unit cell containing several atoms, here we wish to point out that interesting physics does exist when there exist *flat* (dispersionless) band(s). The flat band, or a macroscopic number of degenerate states, has appeared in the condensed matter physics from various contexts.

First one is concerned with the spin magnetism in repulsively interacting itinerant electrons, as exemplified by the Hubbard model. It has become increasingly clear that only at, or possibly around, the singular limit of infinite interaction and infinitesimal doping from a half-filled band does a ferrimagnetism appear. Lieb[3] then pointed out that we can realize a ferromagnetism, for arbitrary strength of the Hubbard U at half-filling, if a bipartite lattice with nearest-neighbor transfers has different numbers, $n_a\neq n_b$, of a and b sublattice sites in a unit cell. In this situation n_a-n_b flat band(s) appear, and the ferromagnetism resides on the flat bands.

This is in accord with the "generalized Hund's coupling,"[4] which dictates that electrons on the Fermi surface should be fully spin polarized for arbitrary U — a macroscopic number of states lying on the Fermi energy will then imply a bulk magnetization. A flat band usually arises when there exist localized eigenstates (or Wannier states) that are mutually disjointed. Remarkably, this does not apply to the flat bands considered here: The most compact state cannot be confined within the unit cell, so that the states have to *overlap* with each other. If one *forces* the localized states to be mutually orthogonal to construct true Wannier states, one ends up with even longer-tailed states. Physically, this is exactly why the spin ferromagnetism emerges when the electron-electron repulsion is turned on: aligned spins can fully exploit Pauli's principle to avoid repulsions.[4,5] This reminds us of the fractional quantum Hall system, where the quantum-liquid ground state is fully spin polarized due to the exchange interaction among the orbitals in a Landau level, a peculiar "flat band" arising itself from magnetic fields. There, orthogonalized Wannier states cannot be constructed either.[6]

The model is extended by Mielke[7] and by Tasaki,[8] which introduce distant-neighbor transfers to prepare flat band(s), on which spins align. Since the flat band is a result of an interference among the nearest-neighbor and more distant transfers, we may call this class the flat band due to interference. By contrast, Lieb's class may be called the flat band due to topology, since only the manner in which the sublattices are interlocked matters.

A class of flat bands has also been conceived in the context of "lateral superstructures" that have superperiods of atomic dimensions in lateral directions. These are envisaged to be realized in organic π-electron materials such as the "long-period graphite" (with period $\sim$ of a few tens of Å), once alleged to be obtained in an attempt to fabricate fullerenes.[9] We can use the group theory[10] to classify all the atomic configurations with a superperiod into semiconducting, semimetallic, and metallic classes. A superperiod, such as super-honeycomb structures, *enforces*, in some classes, the existence of flat bands on top of dispersive ones, which is a systematic realization of Lieb's model.

Now, a natural question is what will happen to the flat bands when a uniform magnetic field is applied. We can in fact expect intriguing phenomena, such as the orbital magnetism as in the "ring-current effect" in fullerenes.[11] In the present paper we reveal from the Hofstadter butterfly for the flat-band systems that the magnetic field leaves the flat bands flat, sandwiched between usual Hofstadter butterflies, for the flat bands arising from topology, while the flat band is developed into a butterfly on its own for the flat bands arising from interference. These imply that not only the spin magnetism but the orbital magnetism are intriguing in flat-band systems.

For the tight-binding model on complex lattices we consider for convenience a rectangular unit cell of $L_x\times L_y$ (which is twice the original unit cell in the case of honeycomb systems). The strength of the magnetic field B applied perpendicular to the system is characterized by $\widetilde{B}\equiv BL_xL_y/\Phi_0=q/p$, where $\Phi_0=h/e$ is the flux quantum and the field is called rational when p,q are integers.

The magnetic field is incorporated in the transfer energy t_{ij} in the usual manner through the Peierls phase as

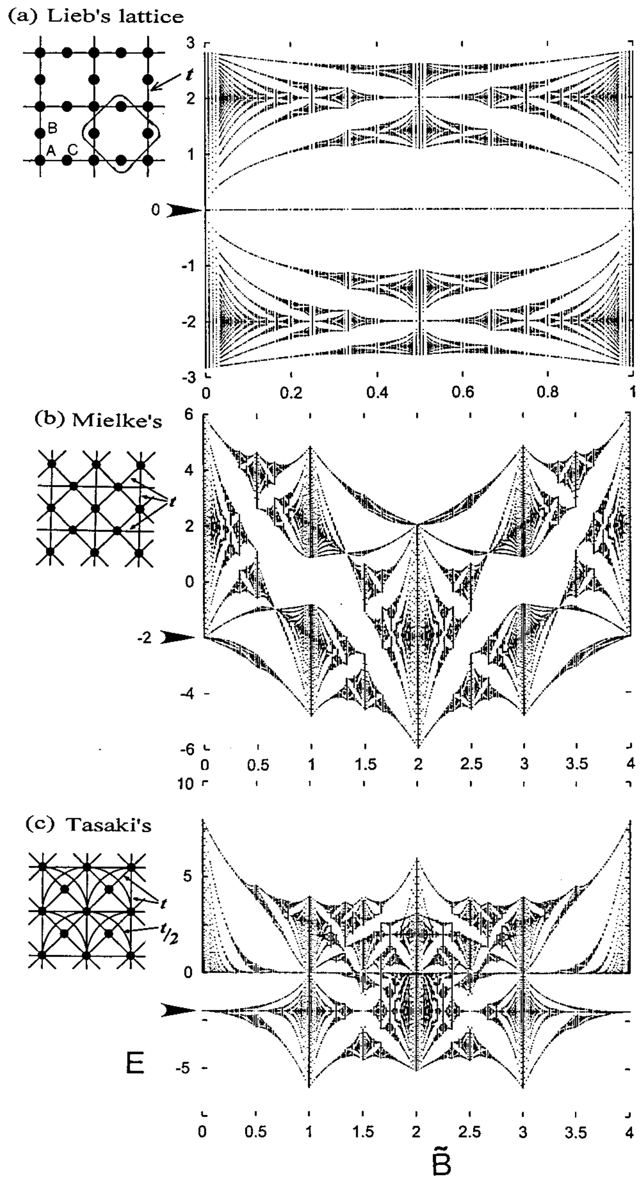

FIG. 1. Hofstadter's diagram (energy spectrum against $\widetilde{B}$) for Lieb's (a), Mielke's (b), and Tasaki's (c) models, whose lattice structures are attached with t etc. being the transfer. Arrows here and figures below represent the position of the flat bands for $B=0$. The spectrum are shown here for $\widetilde{B}\equiv q/p$ with typically $p\leqslant 30$ or $1\leqslant q\leqslant 119$ with $p=120$.

$$t_{ij}\rightarrow e^{i\phi_{ij}}t_{ij}, \tag{1}$$

$$\phi_{ij}=-\frac{2\pi}{\Phi_0}\int_{\mathbf{r}_i}^{\mathbf{r}_j}\mathbf{A}\cdot d\mathbf{r}=-\frac{2\pi B}{\Phi_0}\overline{x_{ij}}\Delta y_{ij}, \tag{2}$$

where the last expression holds for the Landau gauge for the vector potential $\mathbf{A}=(0,\ Bx)$.

In this gauge the phase appears for the transfer involving a shift along x, which repeats itself with a translation of the unit cells along x by $N_{\rm cell}$, where $N_{\rm cell}$ is the smallest N that makes $N(q/p)(\Delta y_{ij}/L_y)$ an integer for all the bonds $\langle ij\rangle$ within or across a unit cell. Thus we can perform a band-structure calculation regarding the $(N_{\rm cell}L_x, L_y)$ system as a new unit cell. Its size depends by construction not only on q/p but also on the atomic configuration in the original unit cell of the superstructure (via Δy_{ij}). Thus the magnetic cell defined here differs from those appearing in the magnetic translation group. The existence of the cell implies that the Brillouin zone will be $N_{\rm cell}$ folded.

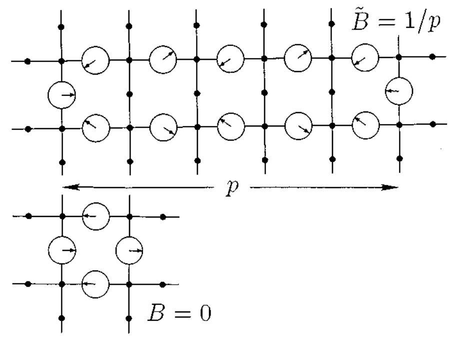

FIG. 2. An example of the $E=0$ ''elongated ring states'' in the Landau gauge, whose length equals p for $\widetilde{B}=1/p(=1/5$ here). A circle represents a finite amplitude, while arrows indicate the phase (which depends on the position of the ring). The inset (left bottom) shows a similar plot for $B=0$. This state is recovered for $p\rightarrow\infty$ as a superposition of the elongated ring states.

Figure 1 displays the Hofstadter butterfly for simple realizations of Lieb's, Mielke's, and Tasaki's models, all assumed to have the square symmetry for simplicity. We can see that, while each dispersive band splits into magnetic minibands, the flat band in Lieb's case remains flat. By contrast the interference-originated flat bands develop into peculiar butterflies as B is increased.

The fact that the topological flat bands can still remain flat in B is analytically shown. There we have only to solve three simultaneous eigenequations for the amplitudes in the unit cell [e.g., ψ_A, ψ_B, ψ_C with A,B,C depicted in Fig. 1(a)]. If we eliminate ψ_B and ψ_C the equation for nontrivial solutions for ψ_A reduces to the corresponding equation for a simple square lattice if we translate $E_{\rm square}$ into E^2-4. On top of these there are N-fold degenerate $E=0$ states that have $\psi_A\equiv 0$, so that we have indeed a flat band with its energy pinned at the original energy that is actually sandwiched by two butterflies mapped via $\pm(E_{\rm square}+4)^{1/2}$. Here N is the number of unit cells, and the atomic level is taken to be $E=0$ (which coincides with E_F when half-filled, i.e., one electron per atom) with $t=-1$.

For $B=0$ the most compact ''Wannier state'' (nonorthogonal as mentioned) on the flat band is depicted in inset of Fig. 2. In quantum chemistry for finite molecules, these states correspond to nonbonding molecular orbitals. Here we have found by inspection that this can be extended to $B\neq 0$ as displayed in Fig. 2. Curiously, B acts to deform the $E=0$ states into ''elongated ring states'' along x (or y) in the Landau gauge, whose length equals p for $\widetilde{B}=q/p$ due to the Peierls phase. This sharply contrasts with the usual Bloch-Landau state having the size of the magnetic length $\propto 1/\sqrt{B}$.

On the other hand, it is not surprising that the flatness is lost even for an infinitesimal B in Mielke's or Tasaki's models, which rely on an exact tuning of the interference. In this case an eigenstate turns from a compact one into a Bloch-Landau state. We can observe some global symmetries in the butterflies such as the following: (i) a full periodicity is ac-

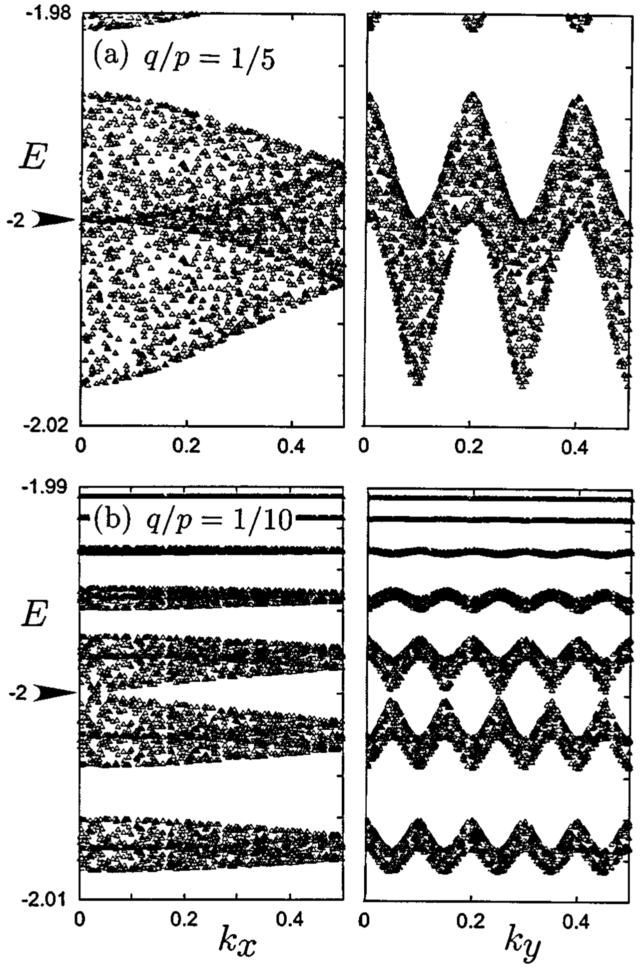

FIG. 3. Typical band structures (projected onto k_x or k_y axis for finite numbers of $\mathbf{k}$'s) are displayed for $\tilde{B}=q/p$ with p even [$q/p=1/10$, (a)] or p odd [$q/p=1/5$, (b)] in Tasaki's model. Note a change in the vertical scale between (a) and (b).

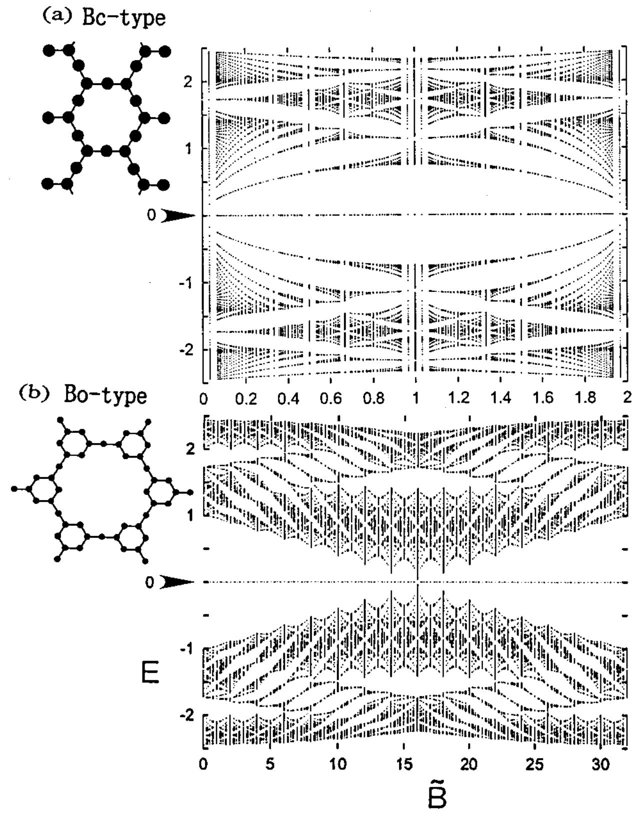

FIG. 4. Hofstadter's diagram for class B_0 (a) or B_C (b) superhoneycomb systems.

complished when the magnetic flux penetrating the smallest loop in the lattice becomes Φ_0 (which is reminiscent of the AB effect in the mesoscopic conductance[12]), (ii) there is a twofold symmetry about $\tilde{B}=1$ for Mielke's model or a mirror symmetry about $\tilde{B}=2$ for Tasaki's.

If we more closely look at the way in which the flat band develops into a butterfly for Tasaki's model in Fig. 3, we find the following. For $\tilde{B}=q/p$ with p even, we have a series of Landau bands (Harper-broadened Landau levels in nonparabolic bands) that have a zero gap at the position, $E_0=-2$, of the original flat band. For an odd p E_0 is a midband point. Usually the anomalous even-odd alternation occurs around the electron-hole symmetric point ($E=0$) in Hofstadter's butterfly for a bipartite lattice: curiously, this occurs here around E_0.

Thus the orbital magnetic moment, $M=-\partial E_T/\partial B$ with E_T being the total energy, becomes anomalous along with the magnetic susceptibility. A specific effect of the delta-function density of states around E_0 spreading both below and above E_0 with B is that the total energy *decreases* when the magnetic flux is introduced if we start from a flat band less than half-filled. This might lead to an orbital ferromagnetism (a spontaneous induction of a network of persistent currents), although it has been pointed out[13] in the context of the flux phase[14] in correlated electrons that a more accurate estimate of energy has to include the diamagnetic shift and the shrinkage of atomic orbitals.

We now turn to superhoneycomb systems, where in the classification by Shima and Aoki a class B_0 (B_C) system has to have, when bipartite, at least three (one) flat band(s) in the gap of semiconducting (semimetallic) bands. To define the classes, we can first note that a unit cell in a honeycomb system may be regarded as comprising two atomic clusters (or ''superatoms''), where the two do not (case A) or have to (B) share an atom. The center of each superatom (a threefold axis) may (case C) or may not (0) coincide with the position of an atom.

The result for the Hofstadter butterfly (Fig. 4) shows that the flat bands remain flat for $B>0$ no matter whether the flat bands are multiple (B_C) or single (B_0) at $B=0$. For the system depicted in Fig. 4(a) the flat band is sandwiched between the butterfly for the simple honeycomb lattice[15] just as in Fig. 1(a), where the only difference is that the butterflies are now mapped via $\pm(E_{\text{honeycomb}}+3)^{1/2}$.

The presumed superstructures are surprisingly stable against the band Jahn-Teller type distortion as seen from the total-energy calculation.[16] For actual fabrication, one possibility would be to polymerize self-aligned organic molecules as realized in the van der Waals epitaxy.[17] Magnetotransport in these systems will be also of interest as in three-dimensional organic materials.[18]

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