Magnetic-breakdown phenomena and recursive band structures

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The magnetic-breakdown (MB) phenomena in the magnetization oscillation in metals are investigated theoretically in order to explain the interference difference oscillation observed in low-dimensional organic conductors, which is prohibited in the often used network model of semiclassical theory for MB. Our full quantum-mechanical theory reproduces various oscillations, including the interference difference oscillation and demonstrates a smooth crossover to the MB orbit oscillation, or MB phenomenon. This is interpreted in terms of the recursive band structures. We point out some pitfalls when analyzing experiments in higher fields.

Magnetic oscillation in metals[1,2] is a direct manifestation of quantum phenomena. The magnetization oscillation known as de Haas-van Alphen effect (dHvA) or magnetoresistance oscillation as Shubnikov-de Haas (SdH) are powerful tools to probe the Fermi-surface (FS) geometry, the conduction electron relaxation time and the effective mass by analyzing the oscillation period or amplitude via the so-called Lifshitz-Kosevich (LK) formula.[1] The LK formula is firmly established for a simple closed FS and underlies "fermilology".

Partly due to exploitation of new materials which are simple enough to allow us to examine the "established" theory valid for idealized situations and partly due to the development of the experimental techniques which lead us to a new previously unexplored domain; higher fields and lower temperatures or purer samples in our context, are confronted with some difficulties in interpreting experimental results in terms of the established theory in connection with the magnetic oscillation phenomena.

Magnetic-breakdown (MB or breakthrough) phenomena are semiclassically understood as arising from electron tunneling through energy gaps formed on the Brillouin zone (BZ) boundaries by a periodic lattice potential. As shown in Fig. 1, the electron motion on a "small" closed α-FS confined within a BZ in low fields becomes extended at higher fields by reconnecting the electron orbits and tracing a "larger" closed β-FS which overlaps each other along k_y via a magnetic breakthrough. This type of the naive semiclassical picture for MB has been formulated and is known as the network model[1,3] and is widely used to analyze experimental data to yield various useful material parameters mentioned above.

Quasi-two-dimensional conductors[4] such as (BEDT-TTF)$_2X$ [or (ET)$_2X$] and (DMET)$_2X$ under perpendicular fields (H) provide a testing ground for reexamining the existing theories because these are relatively clean systems compared to inorganic materials and their FS's are simple enough to allow us to employ a simple tight-binding model. Namely, the FS for (ET)$_2X$ [X=Cu(SCN)$_2$, KHg(SCN)$_4$, etc.,] is a cylinder whose cross section is essentially elliptic as shown in Fig. 1. Depending upon the crystal symmetry the energy gap is formed along $k_y=\pm\pi/b$ [X=Cu(SCN)$_2$], resulting in a closed small FS (α) or no gap is formed (X=I$_3$), the FS being a large closed ellipsoid (β). This FS connected along one direction (k_y) is known as the one-dimensional network model for MB,[1,3] which has been thoroughly studied because of the simplest FS to allow MB.

One of the predictions is a smooth crossover of the MB orbits from the α-orbit oscillation in lower fields to the β-orbit oscillation in higher fields whose characteristic field, or breakdown field $H_0\simeq(mc/e\hbar)(V^2/\varepsilon_F)$ (m is the electron mass, V is the periodic lattice potential and ε_F the Fermi energy). Indeed, several dHvA or SdH experiments on these organic conductors not only confirm several aspects of this semiclassical theory for MB but also reveal important disagreements with it: that is, in higher fields in addition to the predicted MB orbits $\beta, 2\beta, \beta+\alpha, \ldots$, they discover the interference difference orbit oscillation $\beta-\alpha$ whose Fourier spectral intensity is rather strong [see Caulfield *et al.*[5] and Sasaki *et al.*[6] for

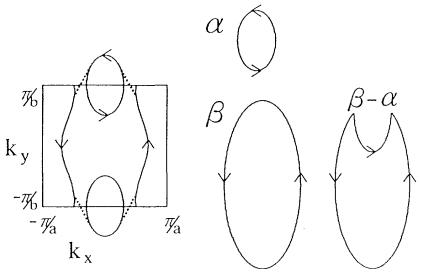

FIG. 1. Calculated Fermi surfaces (FS) under $H=0$, the filling $f=\frac{7}{18}$ and $v=0.09$, consisting of a closed α-FS and a pair of nearly parallel FS's. The dotted lines indicate the breakdown orbit β produced by reconnecting the orbits. The arrows show the direction of the cyclotron motion. Possible breakdown orbits β and $\beta-\alpha$ are drawn. The latter is not allowed in the network model.

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$X=\mathrm{Cu(SCN)_2}$ and Brooks *et al.*[7] for $X=\mathrm{TlHg(SCN)_4}$]. This interference difference orbit is not allowed to exist within the framework of the network model[1] because the direction of the electron motion must be abruptly reversed during a cyclotron motion (see $\beta-\alpha$ in Fig. 1). It is also noted that the overall Fourier spectra of the magnetic oscillations in dHvA or SdH differ from experiment to experiment, which cover different field regions depending on availability of their magnets. This is particularly true for the difference orbit oscillation $\beta-\alpha$ (second largest in Caulfield *et al.*[5] and fourth largest in Sasaki *et al.*[6]). This kind of problem associated with the spectral intensity in magnetic oscillation, or the spectrum instability problem, had already been pointed out long before by Eddy and Stark[8] who measured dHvA on hexagonal Mg. They pointed out on their spectra: (1) the spectral intensity widely differs from oscillation to oscillation, (2) the amplitude variation with H disagrees qualitatively with the network model, and (3) the presence of an interference difference orbit which is not predicted by the network model. These problems have not been solved yet to our knowledge.[9]

In view of the fundamental difficulties inherent in the network model based on semiclassical theory for MB, which is valid for impure samples,[8] with shorter mean free path which mask the long-range quantum coherence, we will try to remedy it by constructing a theory for MB. A principal difficulty of treating the Bloch electrons under H in connection with MB phenomena lies in the fact that there are two conflicting length scales, one coming from a periodic lattice potential and the other from the magnetic length. These are incommensurate with each other in general. This subject, the so-called Hofstadter problem in a broad sense,[10] a task for solving the Harper equation, has been extensively discussed in conjunction with the localization-delocalization transition of the wave functions, the recursive Landau band structure known as a butterfly diagram or the flux state in high-T_c superconductivity. The research front on this problem has greatly advanced in recent years. But it has not been discussed in the context of the magnetic oscillation phenomena to our knowledge. The method developed here turns out to provide a powerful way to solve this problem too. The purposes of this paper are to demonstrate the validity of our method, to provide practical information for the analysis of dHvA or SdH experiments, and finally to urge further careful experiments at lower temperatures and higher fields for purer material to ensure the macroscopic quantum coherence throughout a sample.

Here in order to simulate the FS's in $(\mathrm{ET})_2X$ we employ a two-dimensional tight-binding model Hamiltonian:

$$H=\sum_{k_x,k_y}\left\{-t\left[\cos ak_x+\cos\frac{bk_y}{2}\right]c^{\dagger}_{k_x,k_y}c_{k_x,k_y}+V(c^{\dagger}_{k_x,k_y+2\pi/b}c_{k_x,k_y}+\mathrm{H.c.})\right\},$$

which well describes the essential features of the FS's of this group of materials: $(\mathrm{ET})_2X$.[4] We display the calculated FS for $v=V/t=0.09$ and the electron filling $f=\frac{7}{18}$ in Fig. 1. The periodic potential V gives rise to an energy gap at the BZ boundary $k_y=\pm\pi/b$. This simple one-body problem is equivalent to the situation in the one-dimensional network model mentioned above because the FS for certain electron fillings is extended to the next zone and overlapped each other along k_y direction (see Fig. 1). We notice that this FS situation is realized in κ-$(\mathrm{ET})_2\mathrm{Cu(NCS)_2}$ and α-$(\mathrm{ET})_2\mathrm{TlHg(SCN)_4}$ for $v\neq 0$ and α-$(\mathrm{ET})_2\mathrm{I_3}$ for $v=0$ with three-quarter filling. Namely, a large closed elliptic FS (β) in $v=0$ is modified by the periodic potential into a pair of nearly parallel open FS's and a small closed elliptic FS shown in Fig. 1.

The magnetic field H is introduced by a standard way known as the Peierls substitution scheme;[11] $ak_x\rightarrow ak_x+(ea/\hbar c)A_x=ak_x+(\delta/i)(\partial/\partial k_y), bk_y\rightarrow bk_y$ for the Landau gauge: $\mathbf{A}=(Hy,0)$, where $\delta=(eaH/\hbar c)=(\phi/\phi_0)(2\pi/b)$ with the flux $\phi=abH$ passing through a unit cell $a\times b$ (the unit flux $\phi_0=hc/e$). We obtain

$$H=\sum_{k_x,k_y}\left\{-\frac{t}{2}(e^{iak_x}c^{\dagger}_{k_x,k_y-\delta}c_{k_x,k_y}+\mathrm{H.c.})-t\cos\frac{bk_y}{2}c^{\dagger}_{k_x,k_y}c_{k_x,k_y}+V(c^{\dagger}_{k_x,k_y+2\pi/b}c_{k_x,k_y}+\mathrm{H.c.})\right\},$$

which reduces to the Harper equation in the Hofstadter problem[10] when $v=0$ mentioned above. When the field is commensurate with the underlying lattice, that is, the flux ϕ per unit cell is given by a form: $\tilde{H}=\phi/\phi_0=eabH/hc=q/p$ (p and q are mutually prime). The problem is reduced to diagonalization of a $p\times p$ matrix under periodic boundary condition (we have ignored the spin to avoid additional complications). The total energy $E(\tilde{H})$ per site, which is scaled by t, is evaluated by summing up the eigenvalues from the bottom up to the occupied state under a given electron filling. We calculate the magnetization $M(\tilde{H})$ by differentiating $E(\tilde{H})$ with respect of $\tilde{H}$ (the Bohr magneton $\mu_B=1$). Since $\tilde{H}$ takes only discrete points of rational numbers, we have performed extensive computations by selecting as many rational numbers as possible within feasible computation time.

The resulting energies $E(\tilde{H})$ as a function of $\tilde{H}^{-1}$ are shown in Fig. 2(a). The following should be noted: The distinctive cusps occur at certain fields which satisfies $\tilde{H}=\phi/\phi_0=2f/n$ and $4f/n$ ($n=1,2,3,\ldots$) for the electron filling factor: f because the Landau bands are completely filled at these field values (See later for more detailed discussion on this point). These cusps ultimately lead to the oscillation phenomena. As $\tilde{H}$ decreases, the oscillatory $E(\tilde{H})$ curves are modulated by a different period. This feature is absent in the $v=0$ case where the only single-frequency F_β (and its higher harmonics) appears throughout the covered $\tilde{H}$ region. Since in $v=0$ the FS is a simple ellipsoid (β), no other oscillation than the β orbit is expected. Our result reproduces this simple case.

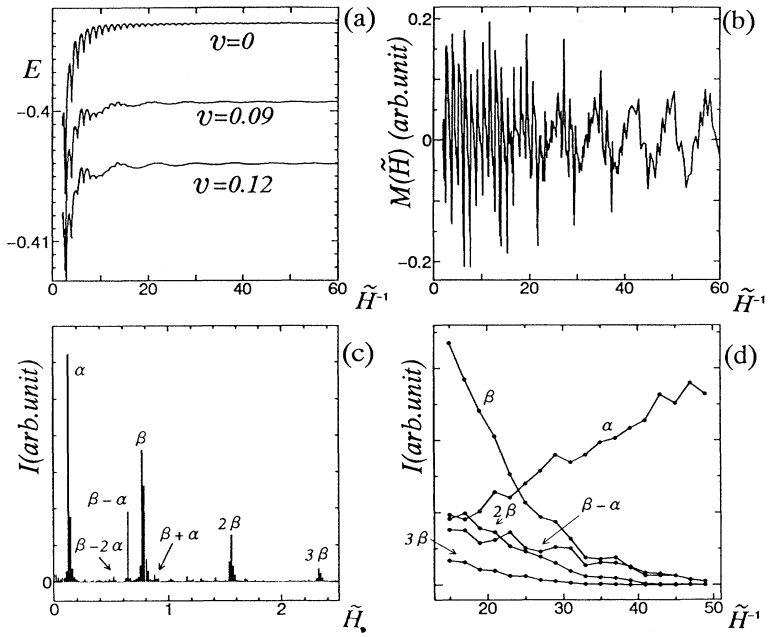

FIG. 2. (a) Total energy $E(\tilde{H})$ per site as a function of $\tilde{H}^{-1}$ for several values of $v(f=\frac{7}{18})$. While a single oscillation is seen in $v=0$, a different rapid period oscillation or the breakdown β oscillation emerges with increasing $\tilde{H}$ at around $\tilde{H}_0^{-1}\sim 25$ for $v=0.09$ and ~ 17 for $v=0.12$. (b) $M(\tilde{H})$ obtained by differentiating E in (a) ($v=0.09$). A smooth changeover of the oscillation periods is evident. (c) Fourier spectrum intensity I_i of $M(\tilde{H})$ taken for whole fields in (b) ($v=0.09$). (d) Fourier spectrum intensity $I_i(\tilde{H})$ ($i=\alpha,\beta,\ldots$) of $M(\tilde{H})$ in (b) taken for a narrow window of $\tilde{H}^{-1}$ and sliding the window. The magnetic breakdown occurs at around $\tilde{H}_0^{-1}\sim 25$. Note the interference difference oscillation $I_{\beta-\alpha}$.

The magnetization $M(\tilde{H})$ which is obtained by numerically differentiating $E(\tilde{H})$ is shown in Fig. 2 (b). Corresponding to the cusps in $E(\tilde{H})$, $M(\tilde{H})$ exhibits a sawtooth behavior. It is evident that a smooth crossover from rapid oscillation to slow oscillation occurs at around $\tilde{H}^{-1}\simeq 25$ for $v=0.09$. This characteristic field called the breakdown field $\tilde{H}_0$ increases upon increasing $|v|$ as inspected from Fig. 2(a) (Compare $v=0.09$ and $v=0.12$).

The overall Fourier spectrum of $M(\tilde{H})$ is shown in Fig. 2(c). There are several definite peaks which can be assigned theoretically. The expected α-orbit, the breakdown β-orbit oscillations and its higher harmonics $2\beta, 3\beta$ are observed. In addition, the interference difference orbit $\beta-\alpha$ which is not allowed in the network model is also seen with substantial intensity, while the expected $\beta+\alpha$ orbit is vanishingly small.

The frequency F for an oscillation is related to the cross-sectional area S of the closed FS in reciprocal space by $F=\phi_0/(2\pi)^2 S$ in the LK formula.[1] This is easily rewritten as $F/H=(S/S_{\rm BZ})/\tilde{H}$ with $S_{\rm BZ}$ being the total area of the first BZ. For the breakdown β-orbit oscillation, $(S_\beta/S_{\rm BZ})/\tilde{H}=2f/\tilde{H}$ (the factor 2 comes from halving of BZ due to v), giving rise to $F_\beta/H=2f/\tilde{H}$, which is the exact relation independent of the system parameters. This relation is in fact satisfied by our calculation, that is, for the filling factor $f=\frac{7}{18}$, $F_\beta/H=\frac{7}{9}\cong 0.778$ in the unit of $\tilde{H}$, coinciding with the frequency shown in Fig. 2(c). As for the α-orbit oscillation, by noting that S_α depends on v, $S_\alpha/S_{\rm BZ}$ is roughly 0.12 as is read off from Fig. 1. This yields $F_\alpha/H=0.12$ in the unit of $\tilde{H}$ which is approximately satisfied as seen from Fig. 2(c). Judging from its frequency, the interference difference oscillation $F_{\beta-\alpha}$ corresponds to the cross-sectional area $S_\beta-S_\alpha$, therefore the orbit area gives rise to a picture shown in Fig. 1 for the $\beta-\alpha$ orbit which is not allowed in the network model mentioned previously. (We are not saying, of course, that this type of classical picture for the $\beta-\alpha$ orbit motion shown in Fig. 1 is correct. This is just for illustration).

The relative intensities $I_i(\tilde{H})$ of these Fourier spectra ($i=\alpha,\beta,\ldots$) under varying $\tilde{H}$, which are obtained by taking the Fourier spectrum for $M(\tilde{H})$ in a certain narrow field interval or a window and by sliding this window, are displayed in Fig. 2(d). We see a smooth changeover of the intensities I_α and I_β, that is, the magnetic breakdown phenomenon. While I_α decreases almost linearly, I_β grows rapidly with $\tilde{H}$. The intensity $I_{\beta-\alpha}$ of the interference difference oscillation behaves similarly to the second harmonic one: $I_{2\beta}$. In a certain field interval $I_{\beta-\alpha}$ could be third next to I_β and I_α, exceeding $I_{2\beta}$. Other intensities such as $I_{\beta+\alpha}$, $I_{\beta-2\alpha}$, and $I_{2\alpha}$ are rather weak or vanishingly small. The fact that the breakdown field H_0 is found to be proportional to $|v|^2$ in our calculations[12] is in agreement with the network model.

Depending upon the field regions where the Fourier spectrum is taken, the relative intensities of various orbit oscillations delicately change as shown in Fig. 2(d). Thus it is not unexpected that different authors[5,6] report different Fourier spectra for κ-$(ET)_2Cu(NCS)_2$, in particular for the $\beta-\alpha$ orbit because they consider different field regions. The result of remarkably different Fourier spectra, of dHvA on Mg,[8] which are taken in only slightly different field regions, is conceivable because in hexagonal Mg much more possible breakdown orbits than the present case exist.[13]

The overall band structures or the distribution of the eigenvalues for various fields are drawn in Fig. 3 in which the Landau levels broaden by a periodic potential into bands, namely the so-called Harper broadening is occurring. Now we can understand why two kinds of the main magnetic oscillations of the α and β orbits arise: From Fig. 3 we can see the two groups of the main gaps or fan

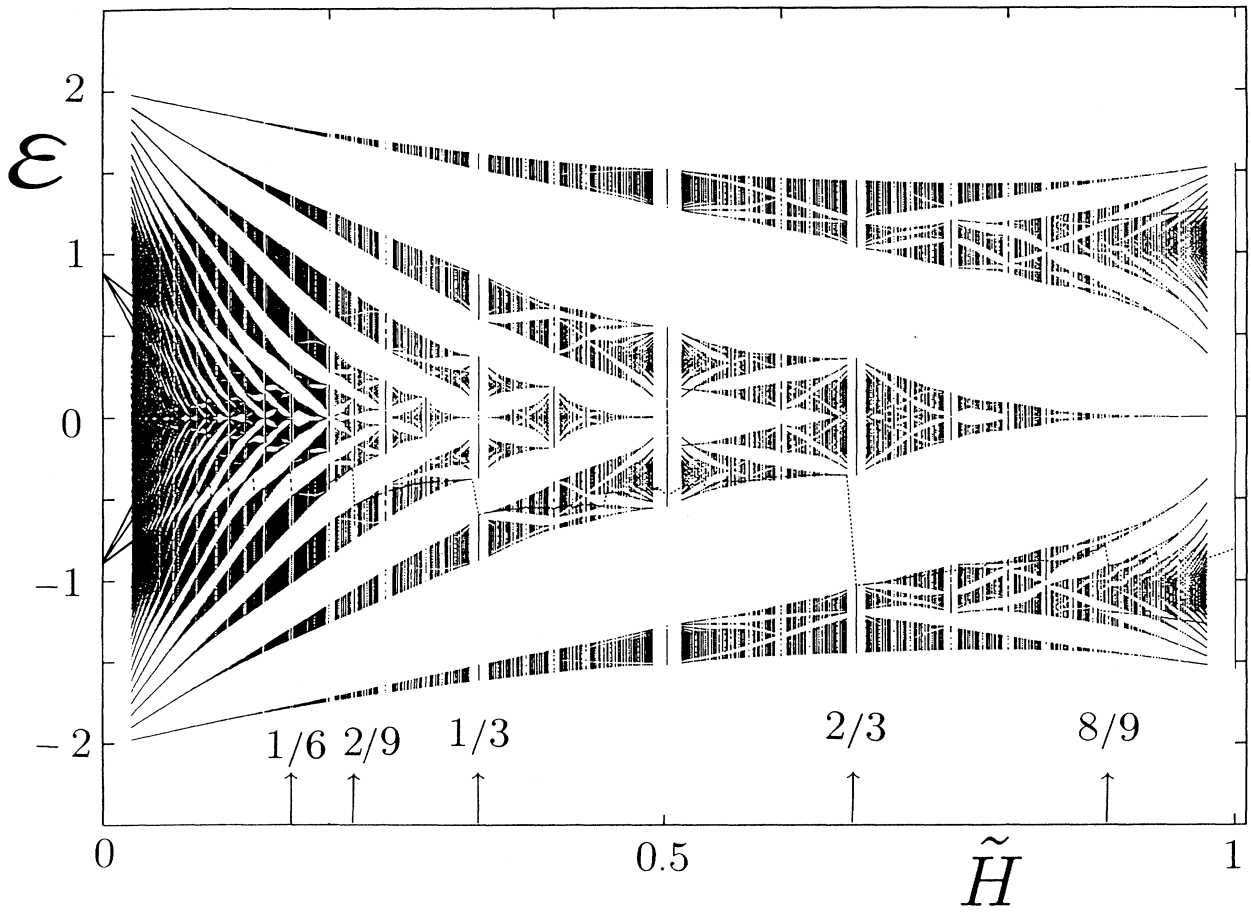

FIG. 3. Overall recursive band structure as a function of $\tilde{H}$ for $v=0.09$. The dotted line indicates the Fermi level ε_F for third filled case $f=\frac{1}{3}$. Arrows denote where the prominent jumps of ε_F occur. From the $\tilde{H}=0$ side two main groups of the gaps start, one from the band edges at $\varepsilon=\pm 2$ and the other from $\varepsilon=\pm 0.8$ indicated by lines.

charts, one emanating from the bottom (or top) of the band edges $\varepsilon=\pm 2$ of the $\tilde{H}=0$ side and the others from the middle $\varepsilon=\pm 0.8$ indicated by the lines in Fig. 3 at which the bottom and top of the α-FS band in $H=0$ are situated. As is shown in Fig. 3 as a dotted line, under a fixed electron number the Fermi level (ε_F) traverses upon increasing $\tilde{H}$ and exhibits series of jumps when crossing these two kinds of the gaps, the former giving rise to β-orbit oscillation and the latter to α-orbit oscillation as is seen shortly.

The overall band is apparently recursive, that is, a similar pattern is repeated many times with different scales. In one-third filling case $f=\frac{1}{3}$, for example, ε_F crosses the larger gaps coming from the bottom precisely at $\tilde{H}=2/3n$ ($n=1,2,3,\ldots$) as seen from Fig. 3 at which the cusps in $E(\tilde{H})$ occur. This leads to the β-orbit oscillation whose period $F_\beta/H=\frac{2}{3}/\tilde{H}$. Moreover, associated with the recursive band structure, ε_F exhibits jumps at $\tilde{H}=4/3n$, $5/3n$, $7/3n,\ldots$ when crossing the subsidiary smaller gaps, yielding the higher harmonics 2β, 3β, $4\beta,\ldots$ (see, for example, a jump at $\tilde{H}=\frac{8}{9}$ in Fig. 3). As mentioned, at these $\tilde{H}$ values the Landau bands are completely filled, resulting in the cusps in $E(\tilde{H})$. It is instructive to compare it with the butterfly diagram[10] for $v=0$: In the latter there is only one kind of gap, coming from the top (or bottom) band edges, giving rise to only the β-orbit oscillation as expected [see Fig. 2(a)]. This corresponds to the fact that in the original square-lattice case of the Hofstadter problem the FS is a simple and isolated closed shape (a square in $f=\frac{1}{2}$) whose area is uniquely determined by f. Further jumps in ε_F associated with the recursive band structure give the higher harmonics 2β, $3\beta,\ldots$ of the β oscillation. The intriguing recursive band structure interestingly enough implies an immediate physical consequence.[12]

It is obvious from Fig. 3 that as $\tilde{H}$ increases, ε_F first crosses the series of α-orbit gaps emanating from $\varepsilon\simeq\pm 0.8$ in lower fields and then meets the β-orbit gaps coming from the band bottom in higher fields. This gives rise to MB phenomenon. Therefore, the larger the α-orbit gaps or v becomes large, the larger the MB field H_0 because the α-orbit gaps persist to higher fields. It is also true that as the filling factor f gets smaller H_0 decreases because ε_F reaches the β-orbit gap-dominated field region in lower fields as understood from Fig. 3.

Urged by the recent observation of the interference

difference oscillation $\beta-\alpha$ in κ-$(ET)_2Cu(SCN)_2$ (Refs. 5 and 6) and β-$(ET)_2TlHg(SCN)_4$,[7] which is prohibited by the often used network model, we have reexamined the magnetic oscillations of dHvA, in particular the magnetic-breakdown phenomena in higher fields. We have presented a general framework which enables us to understand MB in terms of the recursive band structures. Our theory reproduces the standard theory described by the LK formula valid for simple isolated FS's. We have seen that in some aspects the network model based on semiclassical picture agrees with our quantum-mechanical calculations, but fails completely in other aspects. Therefore, it is quite dubious to analyze data in terms of the network model to extract, for example, the effective mass or Dingle temperature. Although we only present a rough sketch for the magnetic oscillations here, we need to go further along this line. Namely, extending to finite temperatures, we will get information on the effective masses by analyzing the temperature dependence of the oscillation amplitudes in combination with the LK formula.

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[11]This scheme itself is under some discussions: Although the absolute values of the calculated Landau bands necessarily agree with first-principles calculation, the relative positions of the bands are accurate. The magnetization oscillations are concerned with the latter. See H. J. Schellnhuber *et al.*, Phys. Rev. Lett. **45**, 276 (1980) and A. S. Alexandrov *et al.*, *ibid.* **66**, 365 (1991).

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