# Magnetic-field-induced transition in quasi-two-dimensional systems

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(Received 12 June 1984)

A model is presented which gives a unique magnetic-field-induced transition from a two-dimensional open-orbit metal to a semimetal. A magnetic field causes the open-orbit electron motion to become progressively more one dimensional, with an effective density of states proportional to $H$ at low field. This leads to a Fermi-surface instability with a transition temperature which varies approximately as $T_c = T_0 \exp(-A/H)$. The resulting gap, which is both temperature and magnetic field dependent, can explain some of the anomalous quantum oscillations and the Hall steps observed in the tetramethyltetraselenafulvalenium compounds [$(TMTSF)_2X$].

The organic conductor perchlorate [$(TMTSF)_2ClO_4$] is a highly anisotropic metal.[1] Its truly unique feature is the anomalous magnetic-field dependence of its low-temperature properties. At low fields the Fermi surface consists of two warped planes and there are only open orbits in the $a$-$b$ plane. Above a temperature-dependent threshold field, $H_k$, quantum oscillations are observed in several experiments suggesting the presence of closed orbits.[2-5] Only the component of $H$ perpendicular to the $a$-$b$ plane is important, indicating an orbital effect in the $a$-$b$ plane. Specific-heat measurements[6] suggest a second-order transition and magnetic resonance[7,8] suggests that the high-field state is a spin-density wave (SDW). Recent Hall measurements[9,10] show some similarity to the quantum Hall effect. However, the quantum oscillations are only quasiperiodic in $1/H$, and the position and magnitude of the Hall steps are highly temperature dependent.

In this work a model is presented which suggests how a magnetic field can cause the electron motion along a two-dimensional open orbit to become progressively more one dimensional and produce an effective density of states which varies linearly with $H$. This leads to a transition temperature for a Fermi-surface instability which has the form $T_c = T_0 e^{-A/H}$. The resulting state, presumably a SDW, has a gap which is both temperature and magnetic field dependent and this may explain many of the anomalies observed experimentally.

The two-dimensional Fermi surface of $(TMTSF)_2ClO_4$ is shown schematically in Fig. 1. A variety of measurements as well as band-structure calculations[11] suggest that the ratio of transfer integrals or bandwidths is $\sim <1{:}10{:}100$ for $t_c{:}t_b{:}t_a$. We take $x$ in the $a$ direction and $y$ in the $b$ direction. For open orbits there is a natural frequency $\omega_c' \sim (eH/m_x c)k_x b$,[12] which replaces the usual cyclotron frequency and corresponds to the frequency with which an electron crosses the Brillouin zone in the $b$ direction in the presence of a magnetic field. When $\hbar\omega_c' > 4t_c$ the system becomes effectively two dimensional. We are effectively two dimensional throughout the interesting magnetic-field regime. If we take the two-dimensional dispersion relation as

$$E(k_x,k_y) = (\hbar^2 k_x^2/2m_x) - 2t_b \cos(k_y b) ,$$

then quasiclassical quantization in a magnetic field corresponds to solving the Schrödinger-type equation:[12]

$$(\hbar^2/2m_x)(\partial^2/\partial x^2)X - 2t_b \cos(k_y b - eHbx/\hbar c)X = EX , \quad (1)$$

where the Landau gauge is chosen. The variable $y$ appears in the equation only as the $k_y$ term in the argument of the cosine. A linear coordinate transformation $x_1 = x - x_0$ (where $x_0 = k_y \hbar c/eHb$) eliminates $k_y$ from Eq. (1). The energy eigenvalues are independent of $k_y$ and depend only on the conjugate variable to $x$, $k_x'$.[12] Thus we are left with a one-dimensional dispersion relation $E(k_x')$ in the appropriate reciprocal space defined by Eq. (1), with reciprocal-lattice vector $G_x' = eHb/\hbar c$. Note that the dispersion relation is one dimensional for any finite value of $H$ (neglecting temperature, scattering, and $t_c$). This nonintuitive mathematical result can be understood by realizing that the quasiclassical motion of the electron on the Fermi surface takes it completely across the Brillouin zone from $-\pi/b$ to $\pi/b$ repetitively. The electron "averages" over all values of $k_y$ and hence its energy cannot depend on $k_y$.

The particular one-dimensional (1D) property of interest is the Fermi-surface instability (Peierls or SDW)

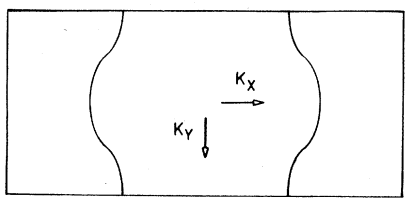


FIG. 1. Schematic diagram of the open-orbit Fermi surface relevant for these materials.

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caused by a large number of degenerate states coupled by the same wave vector. We can define a joint density of states $N_j(E,q)$ as the density of states with energy $E$ coupled by wave vector $q$. In a truly 1D metal all states with energy $E_f$ are coupled by the wave vector $q=2k_f$ across the Fermi surface. Thus $N_j(E_f,2k_f)=N(E_f)=1/E_f$ and there will be a metal-insulator transition at $T_c=T_0\exp[-1/N(E_f)V]$. For the two-dimensional dispersion relation $E(k_x,k_y)$, $N_j(E_f,q)=0$ for all $q$ [although the most favorable $q$ for nesting is $(2k_f,\pi/2)$]. This is consistent with the absence of any density-wave transition for $H=0$.

For small $t_b$ or large $\omega_c'$ Eq. (1) corresponds to a nearly free electron model.[12] For $4t_b/\hbar\omega_c' \ll 1$ the eigenstates near $E_F$ are the usual plane waves with energy $\hbar^2k_x^2/2m$ and density of states $1/E_F$. The system is literally 1D and $N_j \sim 1/E_F$. In this high-field limit $q=(2k_F,0)$. Since $N_j$ is 0 at $H=0$ and $1/E_F$ at high field, the joint density of states has the form $N_j=(1/E_F)F(\hbar\omega_c'/4t_b)$ where $F(x)$ is a smooth function equal to 0 for $x=0$ and saturating to 1 for large $x$. Of interest for the materials under question is the low-field behavior.

For small $\omega_F'=\omega_c'(k_x=k_F)$ the eigenstates of Eq. (1) corresponding to $k_F'$ are a superposition of plane waves centered on $k_F$, separated in $k_x$ by $eHb/\hbar c$, in energy by $\hbar\omega_c'$ and covering an energy range $4t_b$. There are $\sim 4t_b/\hbar\omega_c'$ different plane-wave states comprising the eigenstate $k_F'$. The density of states at $k_F'$ is $\sim 1/E_F$. The fraction of these states which corresponds to the plane-wave state $k_F$ is $\sim\hbar\omega_F'/4t_b$. A distortion at wave vector $q$ couples $k_F$ with $-k_F$. The density of states coupled by $q$ is $(1/E_F)(\hbar\omega_F'/4t_b) \propto H$. Thus $F(x)=x$ for small $x$. Equation (1) can be rewritten as Mathieu's equation and the same result can be obtained from the Fourier transform of the Mathieu functions.

Thus a two-dimensional system with no transition at $H=0$ is unstable in the presence of a perpendicular magnetic field. In the low-field limit the transition temperature will have the form $T_c=T_0e^{-1/N_jV}=T_0e^{-A/H}$. This is similar to the expression obtained in Ref. 13 from Landau-level degeneracy in the case of three-dimensional closed orbits and used in explaining the CDW transition in graphite.[14] In Fig. 2 the experimental data from Ref. 9 are shown to fit this form very well.

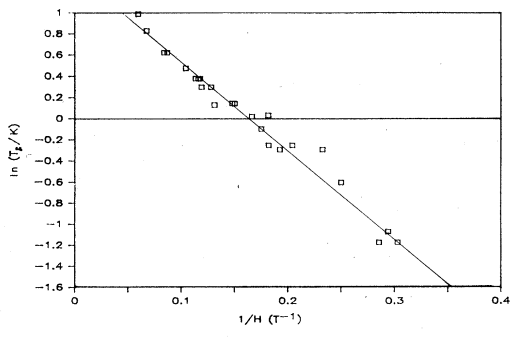


FIG. 2. Temperature dependence of the threshold field, or magnetic field dependence of the transition temperature plotted as $\ln T$ vs $1/H$ from data of Ref. 9.

In real space the transition from two dimensions to one-dimensional behavior is readily seen. An electron in a magnetic field is forced to move on the open-orbit Fermi surface shown in Fig. 1. Its velocity along $x$ is approximately constant. The $y$ velocity oscillates approximately sinusoidally. As the magnetic field is increased the real-space excursion along $y$ is reduced until eventually the electron is localized on a single conducting chain. This occurs when $\hbar\omega_c'=4t_b$ and the system becomes literally one dimensional. The degree of one-dimensionality is thus given by $\hbar\omega_c/4t_b$. Since $t_b \sim 100$ K, and $\hbar\omega_c' \sim (3\ \mathrm{K/T})H$ this ratio is always small. However, as long as the electron motion is limited in one direction and extended in the other the system is one dimensional. This occurs for any finite field, or more realistically, as soon as $\hbar\omega_c'$ is greater than the scattering rate or the bandwidth in the third direction.

The above discussion demonstrates that an open-orbit two-dimensional system with zero joint density of states at $H=0$ attains a finite density of states upon application of a magnetic field and hence is susceptible to a transition related to a Fermi-surface instability. However, the development of the ground state is by no means straightforward. In the absence of a magnetic field one would introduce a potential $v(r)=v_0\cos(\mathbf{q}\cdot\mathbf{r})$ where $q\sim(2k_F,q_y)$ and self-consistently determine $v(r)$ to minimize the free energy. One can approach the problem in two separate ways. The first is to introduce $v(r)$ into Eq. (1) and solve for the eigenstates, fill them up to the correct electron density, and find the form of $v(r)$ which gives the lowest energy. However, this approach violates the quasiclassical approximation used in deriving Eq. (1), namely, that the potentials are slowly varying compared to the original electron wave functions. An additional problem is that the magnetic wave vector $G_x'$ is in general incommensurate with (and much smaller than) $2k_F$ the natural wave vector for $v(r)$. Nonetheless, this approach may be valid for $v_0 \ll \hbar\omega$, i.e., just below the phase boundary for the transition. In this region the induced gap $2v_0$ is a small perturbation on the magnetic energy. There will be oscillations given by the periodicity $nG_x'=q_x\sim 2k_F$. This may be the explanation of the oscillations seen only at high temperatures in Ref. 9.

Unfortunately the above approach breaks down for lower temperatures when $v_0$ becomes comparable to or greater than $\hbar\omega_c'$. Estimating $v_0$ from the transition temperature and studies of other SDW systems, the low-temperature gap is larger than the magnetic energy.

The second approach is to introduce the potential $v(r)$ into the problem before the magnetic field, solve the band structure, find the energy in the presence of the magnetic field, and minimize it by self-consistently varying $v(r)$. In the absence of a magnetic field, the potential $v(r)$ will produce electron and hold pockets with closed orbits and gaps $\sim 2v_0$.[15] The system is a compensated semimetal. When the magnetic field is applied one has conventional Landau quantization of the closed orbits and low-frequency oscillations which correspond to the small orbital area. If the material remains compensated in the presence of the magnetic field then carriers are lost whenever electron and hole Landau levels cross, giving sharp

changes in the Hall effect and magnetoresistance.

The problem with the second approach is that the gaps are never much larger than the magnetic energy $\hbar\omega_c$, so that the possibility of magnetic breakdown is not small. This is certainly true near the phase boundary. Thus the first approach with rapid oscillations is most appropriate at high temperatures and the second approach which gives slow oscillations may be appropriate at low temperatures.

After the original manuscript of this paper had been submitted for publication the author received a copy of the paper by Gor'kov and Lebed.[16] Using a different two-dimensional dispersion relation than presented here, and calculating a generalized susceptibility, they have shown that a two-dimensional open-orbit metal is unstable at any $q$ against a SDW transition due to the increased one-dimensional nature of the electron motion in the presence of a magnetic field. Thus the basic physical ideas are very similar to those presented here. Additionally, they obtain a series of transitions and suggest that the correct $q$ vector is $(2k_F,0)$.

In conclusion, a model has been presented in which the magnetic field effectively increases the one dimensionality of a two-dimensional open-orbit system and hence induces a metal-semimetal transition. Although the ground state and temperature evolution of the system below this transition are as yet unknown, there appears to be a crossover in behavior between the regimes in which the gap is small or large compared to the magnetic energy.

Useful discussions with P. Pincus, T. Witten, T. Lubensky, A. B. Harris, M. Ya Azbel, L. P. Gor'kov, J. F. Kwak, and R. L. Greene are gratefully acknowledged as is research support from the National Science Foundation, Grant No. DMR83-18048.

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