Field-Induced Spin-Density-Wave Instability of the Anisotropic Two-Dimensional Electron System under Lateral Superlattice Potential

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We study the effect of a lateral superlattice potential on the field-induced spin-density-wave (FISDW) instability of the highly anisotropic two-dimensional electron system under magnetic fields. The superlattice with period double the interchain distance causes an oscillatory double splitting of the magnetic energy spectrum. It suppresses the transition into the FISDW subphases with even index numbers. We apply this model to the organic superconductor $(TMTSF)_2ClO_4$ with the anion-ordering superlattice, and give a possible explanation for its anomalous FISDW behavior.

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The organic conductors $(TMTSF)_2X$, where TMTSF denotes tetramethyltetraselenafulvalene and $X=ClO_4$, PF_6, etc., have a very anisotropic electronic structure [1]. It is usually regarded as a highly anisotropic two-dimensional (2D) electron system having a pair of open sheetlike Fermi surfaces (FS's). The field-induced spin-density-wave (FISDW) transitions are the most remarkable phenomena discovered in this system [1]. Most features of the FISDW, particularly those of $(TMTSF)_2PF_6$, are well explained by the "standard model" which deals with the instability of the anisotropic 2D system under magnetic fields [1-4]. However, there still remain unexplained serious problems in the phase diagram of $(TMTSF)_2ClO_4$: the reentrance into the normal phase [5] and a new high-field phase [6]. The "rapid oscillation" (RO) [1,7], possibly a new kind of quantum oscillatory phenomenon, is also an unsolved problem in this system.

The main difference between $(TMTSF)_2ClO_4$ and $(TMTSF)_2PF_6$ is found in the superlattice due to anion ordering (AO) [1]. In $(TMTSF)_2ClO_4$, the orientation of ClO_4 anions orders periodically below 24 K with a period that is double the interchain distance, whereas there is no AO in $(TMTSF)_2PF_6$. The AO superlattice introduces a weak periodic potential in the electron system, and modifies the original FS's by opening gaps. Chang and Maki discussed the effect of the AO on the usual SDW state [8]. Nevertheless, under magnetic field, the AO effect has been believed to be unimportant since electrons easily pass through the AO gap by magnetic breakdown.

A few authors have considered the AO effect as an origin of the anomalous behavior in $(TMTSF)_2ClO_4$ [9-11]. Yan *et al.* first pointed out that the coherent magnetic breakdown causes the oscillatory effect in the same way as the Stark quantum interferometer in ultrapure Mg [10]. They attributed the RO to this interference effect, but it was not clear if this mechanism could explain the RO of the thermodynamic quantities such as the magnetization. Lebed and Bak first studied the FISDW instability under the presence of the AO superlattice [11]. Although they could find the high-field reentrance and the RO, their treatment was not thorough enough because they studied only the $N=0$ subphase by fixing the SDW wave vector.

In this paper, we study the FISDW instability of the anisotropic 2D electron system under a lateral superlattice potential in order to explain the anomalous behavior of $(TMTSF)_2ClO_4$. We take account of the change of the SDW wave vector and consider the modification of the FISDW cascade transitions. Our model is a natural extension of the conventional standard model, but the resulting picture is quite different from that of the Lebed-Bak theory.

We employ the following band model for the anisotropic 2D electron system having open FS's:

$$E(\mathbf{k})=\hbar v_F(|k_x|-k_F)-2t\cos bk_y-2t'\cos 2bk_y\,, \qquad (1)$$

where v_F is the Fermi velocity along the 1D axis (x axis), t and t' ($t\gg t'$) are the effective interchain transfer integrals, and b is the interchain distance. Under the magnetic field $\mathbf{B}=(0,0,B)$, the effective Hamiltonian in the Landau gauge $\mathbf{A}=(0,Bx,0)$ is given by

$$H^0_{\text{eff}}=\hbar v_F\left(\left|-i\frac{\partial}{\partial x}\right|-k_F\right)-2t\cos\left(-ib\frac{\partial}{\partial y}+\frac{beB}{\hbar}x\right)$$
$$-2t'\cos\left(-2ib\frac{\partial}{\partial y}+2\frac{beB}{\hbar}x\right). \qquad (2)$$

Here, we neglected the Zeeman term. The energy spectrum $E^0_{\mathbf{K}}$ and eigenstate $|F^0_{\mathbf{K}}\rangle$ are easily obtained,

$$E^0_{\mathbf{K}} = \hbar v_F(|K_x| - k_F)\,, \tag{3}$$

$$\langle \mathbf{r}|F^0_{\mathbf{K}}\rangle = \frac{1}{\Omega^{1/2}} \exp\left[i\mathbf{K}\cdot\mathbf{r} + i\,\mathrm{sgn}(K_x)\left\{ \frac{2t}{\hbar v_F G}\sin(Gx+bK_y) + \frac{t'}{\hbar v_F G}\sin(2Gx+2bK_y)\right\}\right], \tag{4}$$

where $\mathbf{K}=(K_x,K_y,K_z)$ is the quantum numbers, Ω is the system volume, and $G=beB/\hbar$ is the wave number of the semiclassical open orbit motion in the real space.

We consider a following lateral superlattice potential with a wave vector $\mathbf{Q}=(0,\pi/b,0)$ as a model of the AO in $(TMTSF)_2ClO_4$:

$$U = V\cos(\pi y/b)\,. \tag{5}$$

This periodic potential introduces a new minizone boundary at $|k_y| = \pi/2b$ in the $\mathbf{k}$ space and cuts the original FS's into two pairs of FS's as shown in Fig. 1(a). Also in the $\mathbf{K}$ space, it separates the original first Brillouin zone into two minizones. We limit $\mathbf{K}$ into the first minizone $(-\pi/2b < K_y < \pi/2b)$. The states in the second minizone are represented as $|F^0_{\mathbf{K}+\mathbf{Q}}\rangle$ $(=|F^0_{\mathbf{K}-\mathbf{Q}}\rangle)$. Since U combines $|F^0_{\mathbf{K}}\rangle$ and $|F^0_{\mathbf{K}+\mathbf{Q}+n\mathbf{G}}\rangle$, where n is an integer and $\mathbf{G}=(G,0,0)$, the energy spectrum and eigenstates for the total Hamiltonian $H^0_{\mathrm{eff}}+U$ are obtained approximately:

$$E^{\pm}_{\mathbf{K}} = \hbar v_F(|K_x| - k_F) \pm \Delta,\quad \Delta = VJ_0\left(\frac{4t}{\hbar v_F G}\right), \tag{6}$$

$$|F^{\pm}_{\mathbf{K}}\rangle = (1/\sqrt{2})(|F^0_{\mathbf{K}}\rangle \pm |F^0_{\mathbf{K}+\mathbf{Q}}\rangle)\,, \tag{7}$$

where $J_n(x)$ is the nth Bessel function. The superlattice potential splits the unperturbed spectrum $E^0_{\mathbf{K}}$ into two subbands $E^+_{\mathbf{K}}$ and $E^-_{\mathbf{K}}$ as shown in Fig. 1(b). The splitting energy $2|\Delta|$ oscillates against $1/B$ with the period $\delta(1/B) = \pi v_F be/4t$. This oscillatory splitting results from the coherent magnetic breakdown across the AO gaps in Fig. 1(a).

The energy splitting causes a decrease of the total energy in the same way as the Pauli spin paramagnetism, and this energy gain oscillates, resulting from the oscillation of the splitting. Therefore, the thermodynamic quantities such as the magnetization should show quantum oscillations. It can be shown that the interchain conductivity also oscillates within the framework of the relaxation time approximation [12]. This "paramagnetic" quantum oscillation mechanism gives a possible explanation for the RO observed in the normal phase of $(TMTSF)_2ClO_4$. Using $v_F = 2\times 10^5$ m/s, $t=30$ meV, and $b=0.7$ nm, we can estimate the oscillation period as $\delta(1/B) = 0.0037$ T^{-1}. This is in good agreement with the period observed in the experiments.

In order to study the FISDW instability of this two-subband system, we evaluate the generalized susceptibility $\chi_0(q)$ including no electron correlation following the standard model [2]:

$$\chi_0(\mathbf{q}) = \frac{1}{\Omega}\sum_{\alpha,\beta} |\langle\beta|e^{i\mathbf{q}\cdot\mathbf{r}}|\alpha\rangle|^2 \frac{f(E_\alpha)-f(E_\beta)}{E_\beta - E_\alpha}\,, \tag{8}$$

where $|\alpha\rangle$ and $|\beta\rangle$ are the eigenstates of the system. Using (7) and (4), $\chi_0(\mathbf{q})$ is calculated for $q_x > 0$ as

$$\begin{aligned}\chi_0(\mathbf{q}) = {} & \frac{1}{\Omega}\sum_{m=-\infty}^{\infty} I_{2m}(q_y) \sum_{\mathbf{K}} \frac{f(E^+_{\mathbf{K}}) - f(E^+_{\mathbf{K}+\mathbf{q}+2m\mathbf{G}})}{E^+_{\mathbf{K}+\mathbf{q}+2m\mathbf{G}} - E^+_{\mathbf{K}}} + \frac{1}{\Omega}\sum_{m=-\infty}^{\infty} I_{2m}(q_y) \sum_{\mathbf{K}} \frac{f(E^-_{\mathbf{K}}) - f(E^-_{\mathbf{K}+\mathbf{q}+2m\mathbf{G}})}{E^-_{\mathbf{K}+\mathbf{q}+2m\mathbf{G}} - E^-_{\mathbf{K}}} \\ & + \frac{1}{\Omega}\sum_{m=-\infty}^{\infty} I_{2m+1}(q_y) \sum_{\mathbf{K}} \frac{f(E^+_{\mathbf{K}}) - f(E^-_{\mathbf{K}+\mathbf{q}+(2m+1)\mathbf{G}})}{E^-_{\mathbf{K}+\mathbf{q}+(2m+1)\mathbf{G}} - E^+_{\mathbf{K}}} + \frac{1}{\Omega}\sum_{m=-\infty}^{\infty} I_{2m+1}(q_y) \sum_{\mathbf{K}} \frac{f(E^-_{\mathbf{K}}) - f(E^+_{\mathbf{K}+\mathbf{q}+(2m+1)\mathbf{G}})}{E^+_{\mathbf{K}+\mathbf{q}+(2m+1)\mathbf{G}} - E^-_{\mathbf{K}}}\,,\end{aligned} \tag{9}$$

where

$$I_n(q_y) = \left\{ \sum_{l=-\infty}^{\infty} (-1)^l J_{n-2l}\left(\frac{4t}{\hbar v_F G}\cos\frac{bq_y}{2}\right) J_l\left(\frac{2t'}{\hbar v_F G}\cos bq_y\right)\right\}^2. \tag{10}$$

The four terms in (9) correspond to four combinations of the electron-hole pairing in two subbands, that is, the intrasubband pairings, $(E^+_{\mathbf{K}}, E^+_{\mathbf{K}})$ and $(E^-_{\mathbf{K}}, E^-_{\mathbf{K}})$, and the intersubband ones, $(E^+_{\mathbf{K}}, E^-_{\mathbf{K}})$ and $(E^-_{\mathbf{K}}, E^+_{\mathbf{K}})$. In each term, the summation over $\mathbf{K}$ is taken in the first minizone, and that over m is taken for the possible umklapp processes on $\mathbf{G}$, reflecting the fact that $\mathbf{G}$ works as the reciprocal lattice in the K_x space. Note that the intrasubband pairing allows only the umklapp process with even indices $2m$, and the intersubband one with odd indices $2m+1$.

Equation (9) has multiple local maxima at a fixed magnetic field and temperature. The intrasubband terms show peaks at $q_x = 2k_F + 2MG - 2\Delta/\hbar v_F$ and $q_x = 2k_F + 2MG + 2\Delta/\hbar v_F$ (M is an integer), respectively, and both the intersubband terms at $q_x = 2k_F + (2M+1)G$. These peaks grow with decreasing temperature. Once one peak satisfies the Stoner condition $\chi_0(\mathbf{q}) = 1/I$, where I is the effective interaction constant, the normal electron

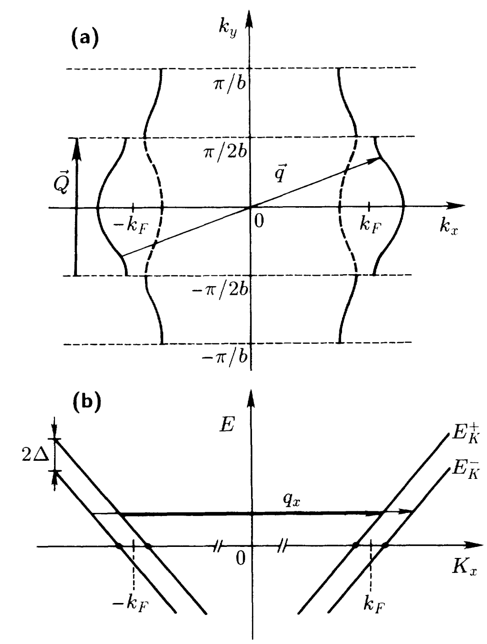

FIG. 1. (a) The schematic FS in the 2D $\mathbf{k}$ space. The dashed curves represent the FS reduced into the first minizone. (b) Magnetic energy spectrum around the Fermi level. The split between two subbands $E_{\mathbf{K}}^{+}$ and $E_{\mathbf{K}}^{-}$ oscillates against $1/B$.

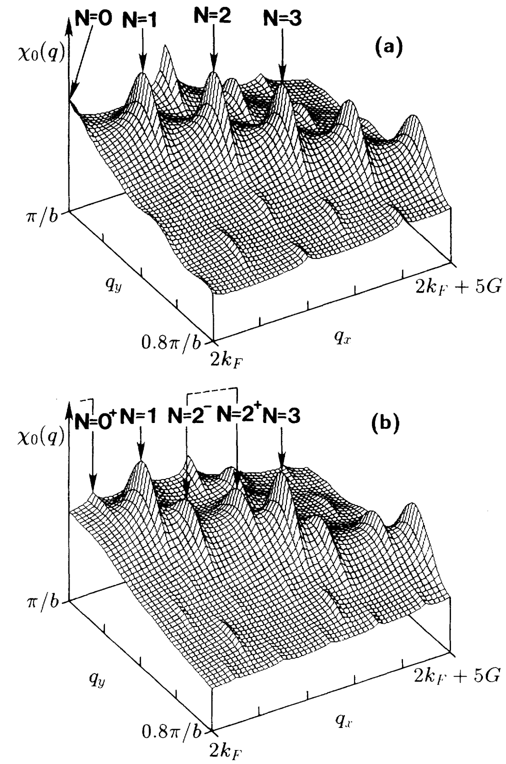

FIG. 2. The q dependence of the susceptibility $\chi_0(\mathbf{q})$ at fixed magnetic field ($\hbar v_F G/4t = 0.03$) and temperature ($k_B T/t = 0.001$) with $t'/t = 0.1$. (a) The case of zero splitting ($\Delta = 0$). (b) The case of finite subband splitting ($2\Delta/\hbar v_F G = 0.5$).

system becomes unstable and undergoes the transition into the SDW phase with a wave vector $\mathbf{q}$.

Therefore, the peaks of $\chi_0(\mathbf{q})$ correspond to the channels into the different FISDW subphases with different $\mathbf{q}$, and the highest peak gives the subphase into which the normal phase really undergoes the transition.

The $\mathbf{q}$ dependence of $\chi_0(\mathbf{q})$ is visualized in Fig. 2. Figure 2(a) shows $\chi_0(\mathbf{q})$ when the splitting energy 2Δ is zero. This is the well-known result of the standard model [2]. $\chi_0(\mathbf{q})$ has peaks at $q_x = 2k_F + NG$ (N an integer), which correspond to the conventional Nth FISDW subphases. Once the splitting energy becomes finite, $\chi_0(\mathbf{q})$ is modified as shown in Fig. 2(b). Each peak with an even index number $N = 2M$ in Fig. 2(a) splits into two small peaks labeled N^- and N^+ at $q_x = 2k_F + NG - 2\Delta/\hbar v_F$ and $q_x = 2k_F + NG + 2\Delta/\hbar v_F$. The transitions into the corresponding subphases, which originate from two kinds of intrasubband pairing, are considerably suppressed compared to that into the original Nth subphase. On the other hand, the peaks with odd index numbers $N = 2M+1$ are not affected by the subband splitting. So, the transitions into the odd-indexed subphases, which originate from the intersubband pairing, show no explicit change even if splitting exists.

Since the subband splitting oscillates periodically against $1/B$, the instability of the even-indexed FISDW subphase also oscillates. The $1/B$ dependence of the $\chi_0(\mathbf{q})$ peaks are shown in Fig. 3. The peaks with odd indices $N = 1$ and 3 show moderate change almost independent of the splitting. The peaks with even indices $N = 0^+$, 0^-, 2^+, and 2^- show oscillatory suppression compared to their envelopes drawn by the dotted curves which show the peak height when the superlattice is absent ($V = 0$).

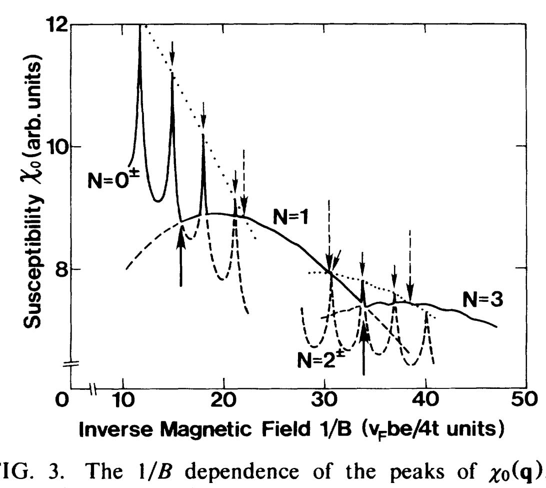

FIG. 3. The $1/B$ dependence of the peaks of $\chi_0(\mathbf{q})$. $V/t = 0.05$, $t'/t = 0.1$, $k_B T/t = 0.001$. Dashed curves show the peaks labeled N and the solid curve shows the highest peak. Dotted curves show the peaks in the case of $V = 0$.

The change of the highest peak means that a transition occurs between the subphases which really appear at low temperatures. In the case of $V=0$, such changes occur at the field positions indicated by dashed arrows in Fig. 3. They give the cascade transitions in the standard model [2]. When the superlattice exists, the main transitions occur between the odd-indexed subphases, for example, between $N=1$ and $N=3$ as indicated by the large arrow at the right. The suppressed even-indexed subphases appear only periodically around the main transitions as shown by the small arrows. This might be observed as the RO. The $N=1$ subphase, which expands to the higher-field region because of the suppression of the $N=0^+$ and 0^- subphases, becomes weak with increasing field. This causes the reentrance into the normal phase. The suppressed $N=0^+$ or 0^- subphase is finally stabilized above the field position indicated by the large arrow at the left. The transition temperature into this high-field phase oscillates against $1/B$. This might give the RO in the high-field region.

Finally, we apply the present picture to $(TMTSF)_2ClO_4$. As the real amplitude V of the AO superlattice potential is unknown, we use the AO transition temperature 24 K as V, so that the amplitude of the oscillatory splitting 2Δ is estimated as 0.4 meV at $B=10$ T. Because this is not negligible compared to the "cyclotron energy" $\hbar V_F G$, which is about 1.4 meV at 10 T, the AO superlattice could cause the effects mentioned above.

We assign the so-called $N=\frac{1}{3}$ subphase [13], which occupies the large region $8<B<27$ T and $T<5.5$ K in the phase diagram, to the $N=1$ subphase. Assuming this, the ratio of the Hall voltage step at the 8-T transition should be 1:3 [13]. The reentrance of the normal phase above 20 T [5] is explained by the suppression of the $N=0$ final subphase. The new high-field semiconducting phase above 25 T [6] is considered to be the suppressed final subphase. The RO in the metallic phase [7] is explained by the paramagnetic quantum oscillation mechanism. In addition, the periodic suppression of the even-indexed subphase could make a large contribution to the observed RO near the FISDW transition.

In conclusion, we studied the FISDW instability of the anisotropic 2D electron system under a superlattice potential. The superlattice with period double the interchain distance causes an oscillatory suppression of the even-indexed FISDW subphases and modifies the phase diagram through the oscillatory splitting of the magnetic spectrum. This gives a possible explanation for the anomalous FISDW features of $(TMTSF)_2ClO_4$ with the AO superlattice.

[1] For $(TMTSF)_2X$ and FISDW, see, for example, T. Ishiguro and K. Yamaji, *Organic Superconductors* (Springer-Verlag, Berlin, 1990), and references therein.

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