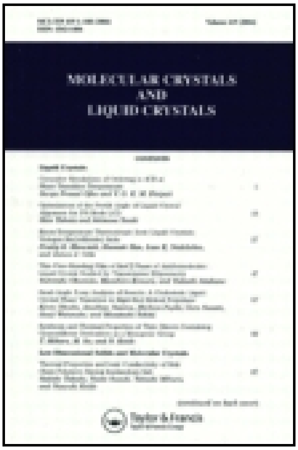

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MAGNETIC BREAKDOWN AND RAPID OSCILLATION PHENOMENA IN LOW DIMENSIONAL ORGANIC CONDUCTORS

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Abstract The rapid oscillation phenomena observed in $(TMTSF)_2X$ and the reentrance phenomena at high fields in $(BEDT\text{-}TTF)_2X$ are considered theoretically by a coherent manner. Both phenomena are understood consistently and coherently due to the magnetic breakdown effect. The rapid oscillation of the magnetization in the former is found to be attributable to the particular Fermi surface reconstructed by the anion and spin density wave orderings.

INTRODUCTION

The quantum magnetic oscillations in the magnetization ($M(H)$) known as the de Haas-van Alphen (dHvA) effect and in the magnetoresistance ($\rho(H)$) oscillation as the Shubnikov-de Haas (SdH) effect are genuine quantum phenomena. These oscillations were described by the so-called Lifshitz-Kosevich (LK) formula in the semi-classical theory based on the Landau quantization[1] which underlies the modern Fermiology. However, it is not applicable when the Fermi surface (FS) becomes complicated a little. It is particularly true when the magnetic breakdown (MB) occurs where FS's are disconnected by a gap. In order to understand the magnetic behaviors in low-dimensional organic conductors[2] $(TMTSF)_2X$ and $(BEDT\text{-}TTF)_2X$, we must properly describe the quantum magnetic oscillations. In $(TMTSF)_2X$ (X=PF_6[3], ClO_4[4,5,6], NO_3[7], ReO_4[8] and AsF_6[9]), the so-called rapid oscillation (RO) periodic as a function of $1/H$ is observed in $M(H)$ and $\rho(H)$ in a wide temperature (T) and magnetic filed (H) regions. The observed RO frequencies (F_{RO}) for all of X's are more or less similar ($\sim$ 260T). The T-dependence ($A_{RO}(T)$) and H-dependence ($A_{RO}(H)$) of the Fourier transformation amplitude (FTA) of the RO have several unexplained features which can not be expected from the standard LK formula of the SdH and dHvA.

The FS of $(TMTSF)_2X$ shown at Fig.1(a) consists of a pair of parallel sheets. The anion ordering (AO) occurs in some X's at low T and the FS is reconstructed (see Fig.1(b); AO vector: $Q_a = (0, \frac{\pi}{b})$ in X=ClO_4 and $(\frac{\pi}{a}, 0)$ in NO_3). In ClO_4 the

AO occurs below the temperature $T_{\rm AO}$ =24K. The RO disappears when the AO is removed by applying a moderate pressure[10]. Therefore, the AO is indispensable for the RO phenomena to occur. Although the Q_a's of ClO_4 and NO_3 are not the same, the frequencies and T-dependences of the $A_{\rm RO}(T)$ of both RO's are very similar. By comparatively studying these similar systems (ClO_4 and NO_3), we might gain a deeper understanding on the RO phenomena.

The characteristics of both RO's are summarized as follows: (1) In ClO_4, the RO of the thermodynamic quantities, e.g., $M(H)$[5], is observed only in the SDW phase, however, the RO in the transport quantity, e.g., $\rho(H)$ is found throughout the SDW and normal state below $T_{\rm AO}$=24K[4,6]. In the normal state $A_{\rm RO}(T)$ in $\rho(H)$ is a decreasing function upon increasing T, whereas in the SDW state $A_{\rm RO}(T)$ has a maximum at $T\sim$ 2K[4,6], which can not be expected from the LK formula. (2) In NO_3, the RO is observed in $\rho(H)$ at low T below the $T_{\rm AO}$=45K. The T-dependence of the $A_{\rm RO}(T)$ has a maximum at $T\sim$ 4K[7]. (3) In addition to the RO another oscillation with the lower frequency $F_{\rm LO}$=67T is also observed in $\rho(H)$[7]. The $A_{\rm LO}(H)$ ($A_{\rm RO}(H)$) decreases (increases) upon increasing H.

Although these RO phenomena were considered theoretically[11], none has succeeded in giving a coherent picture. The purpose of this paper is to give an essence of physics to understand these phenomena coherently through first principles calculations[12].

FORMULATION

The standard model for $(TMTSF)_2X$ is well-known an anisotropic Hubbard Hamiltonian: $\mathcal{H}=\mathcal{H}_0+\mathcal{H}_{int}$, $\mathcal{H}_0=-\frac{1}{2}\sum_{i,j,\sigma}t_{i,j}C^\dagger_{i\sigma}C_{j\sigma}$ and $\mathcal{H}_{int}=\frac{U}{2}\sum_{i,\sigma}n_{i\sigma}n_{i-\sigma}$. where $C^\dagger_{i\sigma}$ is the creation operator of an electron at i site with the spin σ in two-dimensions and U is on-site Coulomb interaction. The Fourier transformation of the hopping integral $t_{i,j}$ yields the standard model band: $\epsilon(k_x,k_y)=-t_a\cos k_xa-t_b\cos k_yb-t'_b\cos 2k_yb$, where $t_a>t_b>t'_b>0$. The energy is measured in units of t_a in the following. The magnetic field applied perpendicular to the (x,y) plane is introduced by the Peierls substitution: $\mathbf{k}\to\mathbf{k}+\frac{e}{\hbar c}\mathbf{A}$, taking the Landau gauge $\mathbf{A}=(Hy,0)$. Under periodic boundary condition, the total energy $E(h)$ of the system is evaluated by diagonalizing the Hamiltonian matrix with a size $p\times p$ for rational fields: $h=\frac{\phi}{\phi_0}=\frac{q}{p}$ ($\phi=abH$: flux passing through the unit cell, ϕ_0: the unit quantum flux. p and q are mutually prime numbers). We obtain the magnetization $M(h)=-\frac{\partial E}{\partial h}$. The AO effect may be expressed as $\mathcal{H}_{\rm AO}=V\sum_{\mathbf{k},\sigma}(C^\dagger_{\mathbf{k}+\mathbf{Q}_a\sigma}C_{\mathbf{k}\sigma}+h.c.)$ with $v=\frac{V}{t_a}$. The FS's reconstructed by the AO potential are displayed by bold lines in Fig.1(b).

The FS for ClO_4 is an ideal situation for the Stark quantum interference (SQI) oscillation for transport properties[1], while these parallel orbits *never* give rise to the thermodynamic oscillation because this does not sustain the Onsager orbital quantization rule. As for NO_3 shown in Fig.1(b), since there exist two closed electron and hole surfaces, SdH and dHvA oscillations should be observed. We consider the effect of the SDW formation on the oscillations whose nesting vector is approximated by $\mathbf{Q}_s = (\frac{\pi}{a}, \frac{\pi}{b})$[14]. In a mean field approximation, $\mathcal{H}_{int}$ is rewritten as $\mathcal{H}_{\rm SDW} = 2\tilde{\Delta}\sum_{\mathbf{k}}(C^{\dagger}_{\mathbf{k+Q}_s\uparrow}C_{\mathbf{k}\downarrow} + h.c.)$. The SDW order parameter $\tilde{\Delta}$ is defined by a self consistent equation: $\tilde{\Delta} = -U\sum_{\mathbf{k}} < C^{\dagger}_{\mathbf{k}\downarrow}C_{\mathbf{k+Q}_s\uparrow} >$. The order parameter Δ ($\Delta = \frac{\tilde{\Delta}}{t_a}$) exerts a periodic potential to further reorganize the electron band. The resulting FS's in $H = 0$ are depicted in Fig.1(c) for both ClO_4 and NO_3. It is now evident that in the FS of ClO_4 there appear possible *closed orbits*, electron and hole pockets, depicted by dotted lines in Fig.1(c) whose area are nearly equal to that in NO_3. It is not difficult to extend the above calculation to finite temperatures. The T-dependence of the order parameter $\Delta(T)$ is assumed to be described by the standard mean field manner, *i.e.*, the BCS theory. However, we regard the ratio $\Delta(0)/T_{\rm SDW}$ as an adjustable parameter because of an experimental fact that the BCS relation $\Delta(0)/T_{\rm SDW} = 1.76$ is often violated in low dimensional conductors[15]. The method of calculations for $M(h,T)$ at finite temperatures is the same as before[13].

RESULTS AND DISCUSSIONS

ClO_4 AND NO_3

Calculated magnetizations ($M(h)$) at $T = 0$ is summarized at Fig.2, where $\Delta = 0$ and $\Delta = 0.03$ correspond to the normal state and SDW state respectively. In ClO_4, at $\Delta = 0$, there is no oscillation in $M(h)$, but at $\Delta = 0.03$, the short and long periodic oscillations are found in $M(h)$. The frequency of the short periodic oscillation corresponds to the MB orbit depicted by the dotted lines in Fig.1(c), which is the usual dHvA oscillation. This frequency agrees with the observed RO's frequency. Namely, the RO in the thermodynamics quantity is the usual dHvA oscillation. Although the FS corresponding to $\Delta = 0$ (Fig.1(b)) does not sustain the Onsager orbital quantization rule, its FS is suitable to give the interference effect due to magnetic field, so that the RO in the transport quantity; $\rho(H)$, can be attributable to the SQI oscillation. In NO_3 at $\Delta = 0$, there is a periodic oscillation in $M(h)$ whose frequency ($f_{\rm RO}$) corresponds to the areas of the closed electron or hole orbits (see Fig.1(b)). At $\Delta = 0.03$ in addition to $f_{\rm RO}$, the oscillation with

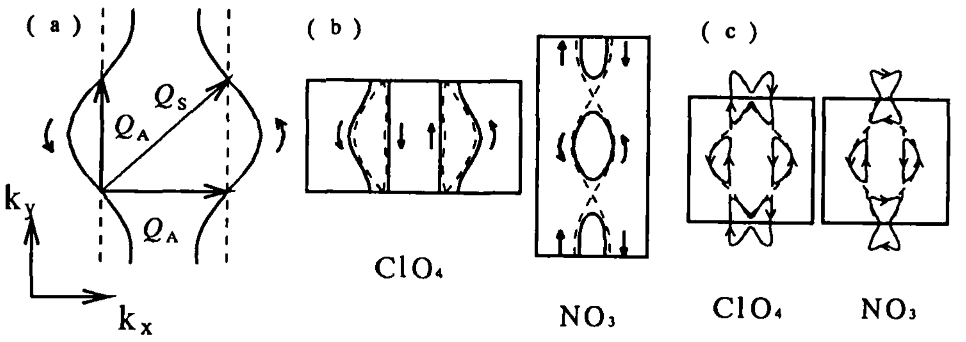

Figure 1: (a) A pair of open FS's ($t_b/t_a = 0.6$ and $t'_b/t_a = 0.2$). $\mathbf{Q}_a$=$(0, \frac{\pi}{b})$ for ClO_4 and $(\frac{\pi}{a}, 0)$ for NO_3. $\mathbf{Q}_s = (\frac{\pi}{a}, \frac{\pi}{b})$. (b) FS's (bold lines) folded by the AO ($v = 0.2$). (c) FS's by the AO and SDW ordering ($\Delta = 0.03$). The closed orbits are blocked by the SDW gaps formed on the crossing points of the dotted lines.

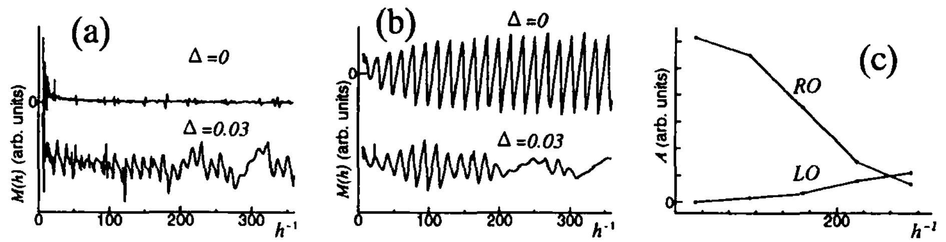

Figure 2: $M(h)$ as a function of h^{-1}. (a) ClO_4. (b) NO_3. (c) In (b) $A_{\rm RO}(h)$ and $A_{\rm LO}(h)$ indicated by dots as a function of h^{-1} ($t_b/t_a = 0.6$, $t'_b/t_a = 0.2$ and $v = 0.2$).

the low frequency ($f_{\rm LO}$) exists in $M(h)$. The closed small electron or hole orbits obtained by the SDW nesting indicated by bold lines in Fig.1(c) corresponds to $f_{\rm LO}$. Moreover, it is seen from Fig.2(c) that $A_{\rm RO}(H)$ ($A_{\rm LO}(H)$) is a smooth increasing (decreasing) function with h, which originates in the MB phenomena.

We investigate the T-dependence of $A_{\rm RO}(T)$ in ClO_4 and NO_3, which are shown in Fig.3 for several values of $\Delta(0)/T_{\rm SDW}$. As for ClO_4 case $A_{\rm RO}(T)$ in Fig.3(b) for $\Delta(0)/T_{\rm SDW} = 5.0$ exhibits non-monotonic behavior near $T_{\rm SDW}$, taking a maximum just below $T_{\rm SDW}$. These non-monotonic T-dependence are understood as follows: The RO comes from the MB orbit blocked by the AO gap and SDW gap as we already pointed out. As $\Delta(T)$ decreases upon increasing T the SDW gap becomes narrowed, thus making the MB easier. This leads to increasing the $A_{\rm RO}(T)$. Namely the T-dependence is governed by the two competing factors: one is the ordinary

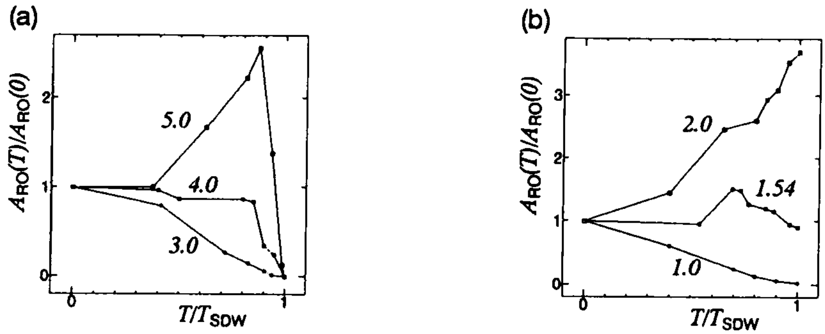

Figure 3: T-dependence of the $A_{\rm RO}(T)$ indicated by dots for several values of $\Delta(0)/T_{\rm SDW}$. (a) ClO_4 and (b) NO_3. $t_b/t_a = 0.6$, $t_b'/t_a = 0.04$, $\Delta(0) = 0.02$ and $v = 0.04$.

temperature reduction due to thermal broadening of the Landau level and the other is the MB enhancement effect due to the gap narrowing. These two factors give rise to the non-monotonic T-dependence. Similarly, as for NO_3 case for $\Delta(0)/T_{\rm SDW} = 1.54$, a non-monotonic behavior in $A_{\rm RO}(T)$ is seen from Fig.3(b), which is caused by the same reason as in ClO_4.

INVERTED SAW TOOTH WAVE FORM IN dHvA

Our calculation reveals a new feature in dHvA. We obtain the inverted saw tooth wave form of $M(h)$ as seen from Fig.2(b) ($\Delta = 0$). This occurs when electron and hole orbits have the almost same area (see the FS for NO_3 in Fig.1(b)). It should be noticed that under this particular FS's situation, the LK formula based on the semi-classical theory with the electron number fixed never yields this inverted wave form.

α-(BEDT-TTF)$_2$ MHg(SCN)$_4$ (M=K, Rb and Tl)

Because of the anomalous T-dependence of $A_{\rm RO}(T)$ mentioned above, the cyclotron effective mass (m^*) cannot be estimated by using the so-called mass plot from the SdH or dHvA experiments. If the MB gap has the T-dependence (the SDW or CDW gaps, etc.), m^* cannot be estimated by using the mass plot. The resulting m^* is not correct even if the FTA is fitted to the LK formula in the narrow T-range. There is recent experimental evidence[16] that this is indeed the case. In α-(BEDT-TTF)$_2M$Hg(SCN)$_4$ having the characteristic kink field ($H_{\rm K}$=24T), Harrison et al.[16] observe that m^* of the small hole pocket (α orbit) monotonically increases from $1.5m_0$ to $2.7m_0$ (m_0 is the bare mass) as a function of H between 18T and 27T.

This behavior is understood as follows: In this system the reentrance transition from the SDW to the normal state occurs at H_K[17]. The α oscillation comes from the MB orbit blocked by the SDW gap. Namely, the enhanced FTA due to the SDW gap closing results in the smaller m^*. In the normal state above H_K, the correct m^* may be estimated for the α orbit. The monotonical increasing of m^* in the intermediate field region originates in the coexistence of the SDW and normal states.

CONCLUSION

We have seen that the RO in $(TMTSF)_2X$ and the reentrance phenomena in $(BEDT\text{-}TTF)_2X$ closely relate to the MB phenomena. As for the former system with $X=ClO_4$, McKernan et al.[18] recently claim the existence of the new SDW phase at still higher fields, where the RO is also found. We need to investigate this region taking into account the MB phenomena theoretically.

REFERENCES

1. D. Shoenberg, Magnetic oscillation in metals (Cambridge University Press: Cambridge, 1984).
2. See for review, T. Ishiguro and K. Yamaji, Organic Superconductors (Springer-Verlag, Berlin, 1990)
3. J. P. Ulmet, *et. al.*, J. Phys. Lett. **46**, L-535 (1985).
4. T. Osada, *et. al.*, Solid State Commun. **60**, 441(1986).
5. X. Yan, *et. al.*, Phy. Rev. **B36**, 1799 (1987).
6. S. Uji, *et. al.*, preprint.
7. A. Audouard, *et. al.*, Phys. Rev. **B50**, 12726 (1994).
8. W. Kang, *et. al.*, Phys. Rev. **B43**, 11467 (1991).
9. A. Audouard,*et. al.*, Synthetic Metals **70**, 751 (1995).
10. H. Shinagawa, *et. al.*, Physica **B201**, 490 (1994).
11. A. G. Lebed, Phys. Rev. Lett. **74**, 4903 (1995). A. G. Lebed and Per Bak, Phys. Rev. **B40**, 11433 (1989). V. M. Yakovenko, Phys. Rev. Lett. **68**, 3607 (1992). K. Maki, Phys. Rev. **B45**, 5111 (1992).
12. Also see, K. Kishigi and K. Machida, Phys. Rev. **B53**, (1996) (in press).
13. K. Machida, *et. al.*, Synthetic Metals **70**, 853 (1995). Phy. Rev. **B51**, 8946 (1995). K. Kishigi,*et. al.*, J. Phys. Soc. Jpn. **64**, 3043 (1995).
14. In the field-induced SDW in ClO_4, $Q_x = 2k_F + N(ebH/\hbar c)$ of the optimal nesting vector $\mathbf{Q}_s = (Q_x, Q_y)$ depends on the subphase numbering N and H (k_F is the Fermi wave number). The ratio of $2k_F = \frac{\pi}{a}$ and $(ebH/\hbar c)$ is an order of $\frac{\phi}{\phi_0} = 10^{-2} \sim 10^{-3}$ in our field region and Q_y is also almost independent of H.
15. G. Grüner, Density Waves in Solids (Addison-Wesley, New York, 1994).
16. N. Harrison, *et. al.*, Phys. Rev. **B52**, (1995) 5584.
17. K. Kishigi and K. Machida, J. Phys. Soc. Jpn. **64**, 3853 (1995).
18. S. K. McKernan, *et. al.*, Phys. Rev. Lett. **75**, 1630 (1995).