Thermodynamics of field-induced spin-density-wave states in Bechgaard salts. II

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Extending the Gor'kov and Lebed approach to more general spin-density-wave {SDW) vectors, we study theoretically the thermodynamics of the field-induced SDW in Bechgaard salts. The quasiparticle spectrum in the SDW is determined. The thermodynamics is identical to that for a BCS superconductor. We predict a cascade of SDW transitions in magnetic fields, which describes the observed phase diagram of the relaxed bis(tetramethyltetraselenafulvalene) perchlorate $[$ (TMTSF)₂ClO₄] extremely well. We calculate also the magnetization in SDW, which again describes the observed magnetization in $(TMTSF)_{2}ClO_{4}$ quite well.

I. INTRODUCTION

The field-induced spin-density-wave (SDW) state in the $(TMTSF)_{2}X$ family of organic-transfer salts (Bechgaard salts) are currently studied both experimentally¹⁻⁵ and theoretically⁶⁻⁹ (TMTSF is tetramethyltetraselena valene).

In an elegant paper Gor'kov and Lebed⁶ (GL) have shown that the field-induced SDW is intrinsic to the quasi-one-dimensional system. Heritier et al ,⁷ considered SOW with more general wave vector. More recently Yamaji 8,9 has shown within a two-dimensional model, where the quasiparticle energy spectrum is given by

$$
E(p) = -2t_a \cos(ap_x) - 2t_b \cos(bp_y) , \qquad (1)
$$

the ground state is described as a series of SDW states with different SDW vectors, while we have shown recent- \rm{ly}^{10} that Eq. (1) describes a cascade of SDW transitions as the magnetic field is decreased. The transition temperature $T_c(H)$ thus obtained describes quite well the observed $T_c(H)$ of (TMTSF)₂ClO₄ by magnetization measurement.⁵

The object of this paper is to study the thermodynamics
of the N th SDW state thus obtained. Earlier one of us¹¹ (hereafter refer to as I) has shown that the thermodynamics of the zeroth SDW is identical to the BCS state. We shall show in the following that within the mean-field approximation, the thermodynamics of all the Nth states are identical to the BCS state. Within the present approximation the phase boundary between the *N*th SDW and the $(N+1)$ th SDW is independent of temperature. Furthermore we obtain a rather simple expression of the magnetization in the Nth SDW, which describes the observe magnetization⁵ in $(TMTSF)_{2}ClO_{4}$ extremely well.

II. GREEN'S FUNCTIONS

As in I, we shall consider the electron Green's function in the Nth SDW state. The Green's function obeys the Gor'kov equation 12

$$
\begin{aligned} [i\omega - \varepsilon(\mathbf{p} - e\mathbf{A}) - \mu H] G_{\omega}(x, x') &+ \Delta(x) F_{\omega}(x, x') = \delta(x - x') , \\ [i\omega - \varepsilon(\mathbf{p} - \mathbf{Q} - e\mathbf{A}) + \mu H] F_{\omega}(x, x') + \Delta^*(x) G_{\omega}(x, x') &= 0 , \end{aligned} \tag{2}
$$

where $G_{\omega}(x,x')$ and $F_{\omega}(x,x')$ are the Fourier transform of the thermal Green's functions with ω the Matsubara frequency defined by

$$
G_{\uparrow}(x,x') = -\langle T[\psi_{\uparrow}(x)\psi_{\uparrow}^{\dagger}(x')] \rangle ,
$$

\n
$$
F_{\uparrow}(x,x') = -\langle T[\psi_{\downarrow}(x)\psi_{\uparrow}^{\dagger}(x')]e^{iQx} \rangle .
$$
\n(3)

We can write down a similar set of equations for $G_1(x, x')$ and $F_1(x, x')$ where μH in Eq. (2) is replaced by $-\mu H$, $Q\rightarrow-Q, \Delta\rightarrow\Delta^*$.

Furthermore the self-consistency equation is given by

$$
\Delta^*(x) = UT \sum_{\omega} F_{\omega}(x, x) , \qquad (4)
$$

where U is the on-site Coulomb potential.

Hereafter we assume⁶ that $\varepsilon(\mathbf{p})$ is given by

$$
\varepsilon(\mathbf{p}) = v \left(|p_x| - p_F \right) - 2t_b \cos(bp_y) - 2t'_b \cos(2bp_y) - 2t_c \cos(cp_z) .
$$
 (5)

As shown by Yamaji⁹ the quasiparticle spectrum (1) is cast in the above form when $(t_b/t_a) \ll 1$, where t_b is given by

$$
t'_b = -\frac{1}{4}t_b^2 \cos(ap_F)/t_a \sin^2(ap_F)
$$
 (6)

we take also $A = (0, Hx, 0)$. Furthermore following Héritier et al ,⁷ we take

$$
\mathbf{Q} = (2p_F + q_x, \pi/b + q_y, \pi/c) , \qquad (7)
$$

where $q_x = -Nk$ and $k = beH$.

Then substituting

$$
G_{\omega}(x,x') = g(x,x') \exp\{i\left[\phi(x) - \phi(x')\right]\}
$$

and

$$
F_{\omega}(x,x') = f(x,x') \exp\{i\left[\phi'(x) - \phi(x')\right]\}\tag{8}
$$

into Eq. (2) we obtain

$$
\begin{cases}\ni\omega + i\upsilon\frac{d}{dx} \\
\frac{\partial}{\partial x} \\
i\omega - i\upsilon\frac{d}{dx}\n\end{cases}\n\begin{cases}\ng(x, x') + \tilde{\Delta}(x)f(x, x') = \delta(x - x')\n\end{cases},\n\tag{9}
$$

with

 $\widetilde{\Delta}(x) = \Delta(x) \exp\{i[\Phi(x) - \Phi(0)]\}$

$$
= \Delta(x)e^{-i\Phi(0)}e^{-iNk(x-x_0)}\sum_{n=-\infty}^{\infty}I_ne^{ink(x-x_0)},
$$
 (10)

where $x_0 = (p_y - \frac{1}{2}q_y)/eH$ and

$$
\phi(x) = v^{-1} \int_0^x du \left[vp_F - \mu H - t (p_y - eHu, p_z) \right],
$$
\n(11)
\n
$$
\phi'(x) = v^{-1} \int_0^x du \left[vp_F - \mu H + vq_x + t \left[p_y - eHu - q_y - \frac{\pi}{b}, p_z - \frac{\pi}{c} \right] \right].
$$

Here

$$
t(p_y, p_z) = -2t_b \cos(b p_y) - 2t'_b \cos(2bp_y) - 2t_c \cos(c p_z)
$$

and (12)

 $\Phi(x)=q_x(x-x_0)+\beta\cos[k(x-x_0)]-\alpha\sin[2k(x-x_0)]$, and 10

$$
I_n = I_n(\alpha, \beta) = i^n \sum_{l = -\infty}^{\infty} J_l(\alpha) J_{n-2l}(\beta)
$$
 (13)

with $J_n(z)$ is the Bessel function,

$$
\alpha = h^{-1}\cos(bq_y), \quad \beta = \eta h^{-1}\sin(\frac{1}{2}bq_y),
$$

\n
$$
h = evbH/2t'_b, \quad \eta = 2t_b/t'_b.
$$
 (14)

As in I the Green's functions $g(x,x')$ and $f(x,x')$ are constructed in terms of eigenfunctions $[u_n(x),v_n(x)]$ as

$$
g(x,x') = \sum_{n} (i\omega - E_n)^{-1} u_n(x - x_0) u_n^*(x' - x_0) ,
$$

$$
f(x,x') = \sum_{n} (i\omega - E_n)^{-1} v_n(x - x_0) u_n^*(x' - x_0) ,
$$
 (15)

where $\psi_n = \begin{pmatrix} u_n \\ v_n \end{pmatrix}$ satisfies

$$
\begin{bmatrix} E_n + iv \frac{d}{dx} & \Delta(x) \\ \Delta^*(x) & E_n - iv \frac{d}{dx} \end{bmatrix} \psi_n(x) = 0 \tag{16}
$$

with

$$
\Delta(x) = \Delta e^{-i\Phi(0)} e^{-iNkx} \sum_{n} I_n e^{inkx} . \qquad (17)
$$

The eigenvalue equation is solved in terms of a plane wave $\psi_n(x) \sim e^{ipx}$. Since the off-diagonal term mixes plane wave e^{ipx} with $e^{i(p + nk)x}$, the energy band splits into a series of subbands as shown in Fig. 1. The energy gaps $2 | \Delta_n |$ at $P = -\frac{1}{2}nk$ are given by

$$
\Delta_n = \Delta I_n \tag{18}
$$

FIG. 1. Quasiparticle energy spectrum in a SDW is shown schematically. The energy spectrum develops a series of energy gaps at $p = \pm p_F + nk/2$, where $n = 0, \pm 1, \pm 2, \ldots$. The Fermi momentum p_F is shifted by Nk/2 so that the Fermi level lies at the center of the largest energy gap.

If we take into account the largest gap Δ_{n_0} exactly and the others perturbatively, the unperturbed energy is given by

$$
E^{\pm}(p) = (n_0 - N)\xi \pm [\xi^2(p) + \Delta_{n_0}^2]^{1/2} , \qquad (19)
$$

where $\zeta = \frac{1}{2} v k$ and

$$
\text{ere } \zeta = \frac{1}{2} v k \text{ and}
$$

$$
\zeta(p) = v (p - p_F + \frac{1}{2} N k) .
$$
 (20)

The details are given in the Appendix. Therefore when $|I_{n_0}| \gg |I_{n_0\pm 1}|, |I_{n_0\pm 2}|, \ldots$, it is most favorable to choose $N = n_0$ for the SDW vector, so that the Fermi surface lies in the middle of the largest energy gap $2 |\Delta_{n_0}| = 2 |\Delta_N|$. In this way the first term in Eq. (19) vanishes and we find

$$
g(x,x') = -\frac{1}{L} \sum_{p} \frac{i\omega + \xi}{\omega^2 + \xi^2 + \Delta_N^2} e^{ip(x-x')}
$$
\n
$$
f(x,x') = \frac{1}{L} \sum_{p} \frac{\Delta_N e^{i\Phi(0)}}{\omega^2 + \xi^2 + \Delta_N^2} e^{ip(x-x')}.
$$
\n(21)

Here we have neglected for simplicity the other energy gapa from the energy spectrum, since their effect is negligible when $|\Delta_{n_0+1}| \ll |\Delta_{n_0}|$. On the other hand when $|\Delta_{n_0\pm 1}| \sim |\Delta_{n_0}|$, a more careful analysis of the effect of the second energy gap is desirable. Finally the gap equation is rewritten as

$$
1 = \tilde{U} |I_N|^2 T \sum_{\omega} \int \frac{dp}{2\pi} (\omega^2 + \xi^2 + \Delta_N^2)^{-1} , \qquad (22)
$$

which is the BCS gap equation where $\tilde{U}= U(bc)^{-1}$.

Therefore we conclude the thermodynamics of the SDW is identical to that of the BCS theory.

FIG. 2. Reduced coupling constant for the *N*th SDW I_N^2 is shown as a function of magnetic field. H_0 is the scaling field given by $H_0 = [t_b/2t_a \cos(ap_F)]^2(eab)^{-1}$.

III. THERMODYNAMICS

Since Eq. (22) is the BCS gap equation for a superconductor, the thermodynamics is identical to a BCS superconductor. First of all the transition temperature is given $\rm{by^{10,13}}$

$$
T_c = 1.14\omega_c e^{-\lambda_N^{-1}}
$$
\n(23)

with $\lambda_N = \lambda |I_N|^2$ and $\lambda = \tilde{U}/\pi v$ at least in the weak coupling limit. We compute $|I_N|^2$ as a function of magnetic field for $\eta = 60$ (which corresponds to $t_a = 2843$ K and $t_b = 265$ K) and plot them in Fig. 2. As the magnetic field decreases $(h < 1)$, the maximum value moves from $|I_0|^2$ to $|I_1|^2$, $|I_2|^2$, ..., in sequence. This gives rise to a series of first-order transitions as the magnetic field is reduced. Recently a qualitatively equivalent result is

FIG. 3. SDW phase diagram is shown. The solid curve shows the transition temperature $T_c(H)$, while vertical thin lines are the phase boundaries between two adjacent SDW's. Broken curves are continuation of the second-order transition lines into the next SDW's.

found by Montambaux et $al.^{14}$ in their numerical analysi of the spin susceptibility $\chi(q,H)$. On the other hand the present theory predicts that the phase boundary between two adjacent SDW states is independent of temperature as shown in Fig. 3. Therefore when the magnetic field is fixed, we stay in a single SDW phase as the temperature is changed. The energy gaps in the vicinity of T_c and at $T = 0$ K are given by¹²

$$
H(T) = \begin{cases} \pi T_c(H)[8/7\zeta(3)]^{1/2}(1 - T/T_c)^{1/2} & (24) \\ \end{cases}
$$

$$
\Delta(H,T) = \left| \frac{\pi}{\gamma} T_c(H) \simeq 1.76 T_c(H) \right|, \tag{25}
$$

respectively, where $\zeta(3)=1.202...$ is the zeta function and $\gamma = 1.78...$ is the Euler constant.

Similarly the free energy is given by

$$
\Delta F = F_{\text{SDW}} - F_n = \begin{cases} -N_0 \frac{(2\pi T_c)^2}{7\zeta(3)} (1 - T/T_c)^2, & \text{for } T \simeq T_c \\ \begin{bmatrix} 1 & \zeta(T_c) \end{bmatrix} \end{cases}
$$
 (26)

$$
=F_{\text{SDW}}-F_n = \left\{ N_0 \left[\frac{1}{3} (\pi T)^2 - \frac{1}{2} \Delta^2 - (2\pi \Delta^3 T)^{1/2} \left(1 + \frac{15}{8} \frac{T}{\Delta} \right) e^{-\Delta/T} \right], \text{ for } T \ll T_c \ . \tag{27}
$$

The free energy at $T = 0$ K (the ground-state energy) is calculated as a function of magnetic fields, which is shown in Fig. 4. The result is quite comparable to the one obtained by Yamaji⁹ for parameters somewhat different from ours.

Perhaps of great interest is the excess magnetization in SD%, which is given by

$$
M = -\frac{\partial}{\partial H}(\Delta F)
$$

= $N_0 \Delta^2(H, T) \lambda_N^{-1} \left[2 \frac{d}{dH} |I_N| \right] |I_N|^{-1}$. (28)

We note that $M(T)/M(0)$ is a universal function of

FIG. 4. Ground-state energy is shown as function of magnetic field.

 $T/T_c(H)$,

$$
M(T)/M(0) = [\Delta(H, T)/\Delta(H, 0)]^{2}.
$$
 (29)

This universal function is shown in Fig. 5. Furthermore we have computed $M(H,0)$ as a function of magnetic fields, which is shown in Fig. 6. Again the magnetization is quite similar to the one due to $Yamaji.⁹$ However, in our calculation the maximum in M for SDW with small N occurs before the transition into the subsequent SDW with larger N unlike Yamajis's result. This feature as well as the relative magnitude of peaks in the magnetization resemble more closely the observed magnetization⁵ in $(TMTSF)_2CIO_4$ at 225 mK. However, the present

FIG. 6. Magnetization at $T = 0$ K is shown as a function of magnetic field.

theory predicts slower decrease in the magnetization in the higher SDW states than the observed magnetization.

Furthermore the magnetization appears to never cross zero for $N \leq 8$ in contrast to the observation. In order that the magnetization takes negative values, $T_c(H)$ should have local maxima, which appears never to happen for $\eta=60$. For a smaller value of η (e.g., $\eta=5$) we have local maxima in $T_c(H)$ [and in $(I_N)^2$] as shown in Fig. 7. However, this choice gives $H_0 \approx 1440$ T within the present model, which is certainly incompatible with the experiment on $(TMTSF)_2ClO_4$.

Also for comparison with the experimental result we have to displace the external magnetic field slightly (-3) T). Therefore we may conclude that the present model describes the qualitative features of the phase diagram as well as magnetization quite well. However, for a quantitative description of the experiment further improvement of the model is clearly desirable.

FIG. 5. Magnetization $M(T)$ follows universal curve as shown here.

FIG. 7. Field dependence of I_N^2 for $\eta = 5$ (corresponding to $t_b/t_a \simeq 1$.

IV. CONCLUDING REMARKS

Extending the method introduced by Gor'kov and Lebed, 6 we have studied the thermodynamics of fieldinduced SD%' in Bechgaard salts. Assuming that the quasiparticle energy spectrum is described by a twodimensional tight binding model given by Eq. (1), we determine the SDW Green's function within mean-field approximation. We have shown that a cascade of SDW states with different SDW vectors are induced in high magnetic fields, which reproduces the observed SDW phase diagram. We find that if we identify the observed phase boundary near $H \approx 10$ T in (TMTSF)₂ClO₄ with the boundary between the $N = 0$ SDW and $N = 1$ SDW, this provides a strong constraint to the present model. This constraint is given as $H_0 \approx 10$ T, where

$$
H_0 = \frac{1}{2} (t_b / t_a)^2 / eab
$$
 (30)

Since a, b as well as t_a are well known, Eq. (30) implies that $t_b/t_a \approx 10^{-1}$.

We have shown also that the present model describes not only the phase diagram but also the magnetization quite well. However, at lower fields the present model predicts higher transition temperature and higher magnetization than those observed experimentally. Furthermore, the present model predicts the phase boundary between adjacent SDW's independent of temperature in contrast to the observation. Therefore some improvement of the model is clearly desirable.

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APPENDIX: ENERGY SPECTRUM OF EQ. (16)

Here we solve Eq. (16) approximately with $\Delta(x)$ given by Eq. (17). Since $\Delta(x)$ is periodic, we look for the solution in the form

$$
u_p(x) = L^{-1/2} e^{ipx} \sum_l a_l e^{ilkx},
$$

$$
v_p(x) = L^{-1/2} e^{ipx} \sum_l b_l e^{ilkx}.
$$
 (A1)

Here L is the length of the system in the x direction. Let $\widetilde{b}_l = e^{-i\Phi(0)}b_l$ and $\Delta_n = \Delta I_n$. Then substituting (A1) into Eq. (16), we obtain equations for a_l and b_l ,

$$
(E - \xi - 2l\xi)a_l + \sum_n \Delta_n \tilde{b}_{l+N-n} = 0,
$$

$$
\sum_n \Delta_n^* a_{l-N+n} + (E + \xi + 2l\xi)\tilde{b}_l = 0,
$$
 (A2)

where $\xi = vp$ and $\zeta = \frac{1}{2}vk$.

If Δ_{n_0} is the largest among Δ_n 's, it is reasonable to take into account Δ_{n_0} exactly and to treat other Δ_n 's perturbationally. For this reason we rewrite the second equation into account Δ_{n_0} exactly and to treat other Δ_n 's perionally. For this reason we rewrite the second equal of (A2), replacing *l* by $l + N - n_0$, as

$$
\sum_{n} \Delta_{n}^{*} a_{l+n-n_{0}} + [E + \xi + 2(l+N-n_{0})\zeta] \widetilde{b}_{l+N-n_{0}} = 0 \text{ and } \sin \zeta
$$

(A3) (l = 0,1,2)

Or we write more compactly

$$
M\cdot \widetilde{\psi} = 0 \tag{A4}
$$

where

$$
\widetilde{\psi}_l = (a_l, \widetilde{b}_{l+N-n_0}), \qquad (A5)
$$

$$
M_{l,l} = \begin{bmatrix} E - \xi - 2l\xi & \Delta_{n_0} \\ \Delta_{n_0} & E + \xi + 2(l + N - n_0)\xi \end{bmatrix}, \quad (A6)
$$

$$
M_{l,l+j} = \begin{bmatrix} 0 & \Delta_{n_0-j} \\ \Delta_{n_0+j}^* & 0 \end{bmatrix}, \text{ for } j > 0
$$
 (A7)

$$
M_{l,l-j} = M_{l-j,l}^{-1}.
$$

$$
M_{l,l-j}=M_{l-j,l}^{+}.
$$

 $M_{l,l}$ is diagonalized by rotating $\tilde{\psi}_l$ into $U_l^{-1}\tilde{\psi}_l$ with

$$
U_l = \begin{bmatrix} u_l & v_l \\ -v_l & u_l \end{bmatrix}
$$
 (A8)

and

$$
u_{l} = \left(\frac{1}{2}\left\{1 + \xi_{l}(p)/[\xi_{l}^{2}(p) + \Delta_{n_{0}}^{2}]^{1/2}\right\}\right)^{1/2},
$$

\n
$$
v_{l} = \left(\frac{1}{2}\left\{1 - \xi_{l}(p)/[\xi_{l}^{2}(p) + \Delta_{n_{0}}^{2}]^{1/2}\right\}\right)^{1/2},
$$

\n
$$
\xi_{l}(p) = \xi + 2\left|l - \frac{n_{0} - N}{2}\right|\xi.
$$
 (A9)

Then we will have

$$
\widetilde{M}_{l,l} = U_l^+ M_{l,l} U_l = \begin{bmatrix} E - E_l^+(p) & 0 \\ 0 & E - E_l^-(p) \end{bmatrix} \quad \text{(A10)}
$$

with

$$
E_I^{\pm}(p) = (n_0 - N)\xi \pm [\xi_I^2(p) + \Delta_{n_0}^2]^{1/2} , \qquad (A11)
$$

when $p\neq0$ or $p\neq\frac{1}{2}k$, these eigenvalues are nondegenerate and a perturbative analysis gives second-order corrections in Δ_n 's ($n \neq n_0$) to $E_l^{\pm}(p)$. On the other hand at $p = 0$ or $\frac{1}{2}k$ energy gaps will develop where two energy spectra intersect. When two branches of the energy spectrum $E_l^{\sigma}(p)$ and $E_m^{\sigma}(p)$ ($\sigma = +, -$) intersect each other at $p = 0$ or $p = \frac{1}{2}k$ then the spectrum in the vicinity of these points is given by

$$
E(p) = \frac{1}{2} [E_l^{\sigma}(p) + E_m^{\sigma}(p)]
$$

$$
\pm \{\frac{1}{4} [E_l^{\sigma}(p) - E_m^{\sigma}(p)]^2 + |\Delta_{l,m}^{\sigma}|^2\}^{1/2}, \quad (A12)
$$

where

$$
\Delta_{l,m}^{\sigma} = (U_l^+ M_{l,m} U_m)_{\sigma\sigma} \tag{A13}
$$

In particular at $p=0$, $E_l^{\sigma}(p)$ and $E_{-l}^{\sigma}(p)$ with $l = 1, 2, 3, \ldots$ intersect and

1,2,3,... intersect and
\n
$$
|\Delta_{l,-l}^{+}| = |u_{l}v_{-l}\Delta_{n_{0}+2l}^{+} + v_{l}u_{-l}\Delta_{n_{0}-2l}^{*}| \simeq |\Delta_{n_{0}+2l}^{+}|,
$$
\n(A14)
\n
$$
|\Delta_{l,-l}^{-}| = |u_{l}v_{-l}\Delta_{n_{0}-2l}^{*} + v_{l}u_{-l}\Delta_{n_{0}+2l}| \simeq |\Delta_{n_{0}-2l}^{-}|,
$$
\n(A15)

and similarly at $p = \frac{1}{2}k$, $E_l^{\sigma}(p)$ and $E_{-l-1}^{\sigma}(p)$ $(l = 0, 1, 2, ...)$ intersect and we have

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$$
|\Delta_{l,-l-1}^{+}| = |u_{l}v_{-l-1}\Delta_{n_{0}+2l+1}+v_{l}u_{-l-1}\Delta_{n_{0}-2l-1}^{\ast}|
$$

\n
$$
\simeq |\Delta_{n_{0}+2l+1}|
$$
\n(A16)

$$
|\Delta_{l,-l-1}^{-}| = |u_{l}v_{-l-1}\Delta_{n_{0}-2l-1}^{*} + v_{l}u_{-l-1}\Delta_{n_{0}+2l+1}|
$$

\n
$$
\simeq |\Delta_{n_{0}-2l-1}|
$$
\n(A17)

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