

Antiferromagnetism in organic conductors

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We obtain the temperature dependence of the spin susceptibility, the spin-flop field, and the antiferromagnetic resonance frequency in a model with overlapping parts of the quasi-one-dimensional Fermi surface and a weak anisotropy of the spin interaction. We show that the magnetic properties are the same as those obtained in a model with localized spins only at $T = 0$. We compare the results with experiments.

1. INTRODUCTION

A new class of organic substances, called Bechgaard salts, which are the TMTSF and TMTTF salts,¹ were synthesized in 1979. In contrast to other quasi-one-dimensional compounds many of them disclose an antiferromagnetic state—a spin density wave (SDW) at temperatures $T < 10$ to 14 K. Below the transition point the substance considered possess all the properties of an antiferromagnetic: anisotropy of the magnetic susceptibility, spin-flop field, antiferromagnetic resonance frequency.^{2–4}

The Fermi surface in Bechgaard salts consists of two open sections with an insignificant transverse dispersion.⁵ The electron energy near the Fermi surface is

$$\xi^{(\pm)}(\mathbf{p}) = \pm v_F(p_{\parallel} \mp p_F) + 2t_b \cos \mathbf{p}\mathbf{b}^* + 2t_c \cos \mathbf{p}\mathbf{c}^*. \quad (1)$$

It was noted in Ref. 6 that the spectrum (1) has a degeneracy connected with the possibility for two sections of the Fermi surface to overlap:

$$\xi(\mathbf{p} + \mathbf{Q}_0) = -\xi(\mathbf{p}), \quad \mathbf{Q}_0 = (2p_F, \pi/b^*, \pi/c^*). \quad (2)$$

In that sense it is analogous to the spectrum of the Keldysh-Kopaev model with embedded electron and hole Fermi surfaces.⁷ The phase transition occurring at a rather low temperature is the pairing of an electron and a hole near different parts of the Fermi surface which lifts the energy degeneracy (2). The order parameter is the quantity

$$\sum_{\mathbf{p}, z} \langle \psi_{\alpha}(\mathbf{p}, z) \psi_{\beta}^+(\mathbf{p} + \mathbf{Q}, z) \rangle.$$

For a number of reasons (*vide infra*) the vector \mathbf{Q} in this expression cannot be the same as the vector \mathbf{Q}_0 which would give exact overlap of the Fermi surface. Sonin⁸ noted in his review paper that an order parameter of the form $\Delta \delta_{\alpha\beta}$ corresponds to a structural instability and $\Delta \sigma_{\alpha\beta}$ to a spin density wave (σ are Pauli matrices).

Earlier one used for the interpretation of the experimental results on the magnetic structure of the compounds considered formulae borrowed from the localized spin model (see, e.g., Ref. 2). As the Bechgaard salts have a high conductivity above the antiferromagnetic ordering temperature, this model does clearly not correspond to the true physical picture as was noted in Ref. 2. Moreover, the model of Ref. 2 does not allow us to obtain the temperature-dependence of the calculated quantities. An evaluation of the magnetic properties of Bechgaard salts in a model with unlocalized electrons is therefore of great interest.

We choose for a comparison of the results obtained with

experiments the compounds $(\text{TMTSF})_2\text{AsF}_6$ (Refs. 2 and 3) and $(\text{TMTTF})_2\text{Br}$ (Ref. 4) in which the main set of quantities which characterize the antiferromagnetic structure were measured.

2. GROUND STATE

The term

$$\psi_{\alpha}^+(\mathbf{p}_+') \psi_{\beta}^+(\mathbf{p}_-') K_{\alpha\beta, \gamma\delta}(\mathbf{p}_+' , \mathbf{p}_-' , \mathbf{p}_-, \mathbf{p}_+) \psi_{\gamma}(\mathbf{p}_-) \psi_{\delta}(\mathbf{p}_+), \quad (3)$$

in the electron interaction operator is important for the formation of a SDW; here $p_{+\parallel}, p_{+\parallel}' \sim p_F; p_{-\parallel}, p_{-\parallel}' \sim -p_F$. Below the transition temperature there appear anomalous mean values relating the wavefunctions of electrons near different parts of the Fermi surface. We can distinguish in the kernel of (3) terms of a Coulomb provenance, exchange terms, and terms with a weak spin anisotropy. The latter is caused by small spin-orbit and dipole-dipole interactions. It was noted in Ref. 9 that the main role in Bechgaard salts in the magnetic anisotropy energy is apparently played by the dipole-dipole interaction. For the sake of simplicity we replace the expression for the screened Coulomb interaction in the momentum representation by a constant (in the weak coupling model the results do not change because of this). We can thus write

$$K_{\alpha\beta, \gamma\delta}(\mathbf{p}_+' , \mathbf{p}_-' , \mathbf{p}_-, \mathbf{p}_+) = \delta_{\alpha\delta} \delta_{\beta\gamma} \Lambda + K_{\alpha\beta, \gamma\delta}^{\text{anis}}(\mathbf{p}_+' , \mathbf{p}_-' , \mathbf{p}_-, \mathbf{p}_+). \quad (4)$$

The equations for the Green functions have the form

$$G_{\alpha\beta}^{++}(iz, \mathbf{p}) = \tilde{G}_{\alpha\beta}^{++}(iz, \mathbf{p}) - \tilde{G}_{\alpha\gamma}^{++}(iz, \mathbf{p}) \Delta_{\gamma\delta}^{+-} G_{\delta\beta}^{--}(iz, \mathbf{p}), \quad (5)$$

$$G_{\alpha\beta}^{--}(iz, \mathbf{p}) = -\tilde{G}_{\alpha\gamma}^{--}(iz, \mathbf{p}) \Delta_{\gamma\delta}^{-+} G_{\delta\beta}^{++}(iz, \mathbf{p}).$$

In this case the self-consistency condition is satisfied

$$\Delta^{-+} = (\Lambda \delta_{ik} + \lambda_{ik}) \sigma_i \frac{1}{2} \text{Sp} \left[\frac{T}{(2\pi)^3} \sum_z \int \sigma_k G^{-+}(iz, \mathbf{p}) d^3p \right]. \quad (6)$$

One sees easily that the equations are invariant under the multiplication of the order parameter by a phase factor. The leeway in the choice of phase corresponds to the possibility of a spatial shift of the incommensurate SDW. In the case of strong commensurability 1:2 states with wavevectors \mathbf{Q} and $-\mathbf{Q}$ correspond to the same quasimomentum and, hence, G^{+-} and G^{-+} are the same. There is then no leeway in the choice of phase. We write the order parameter in the form $\Delta^{-+} = \sigma(\Delta' + i\Delta'')$, where Δ' and Δ'' are real. It follows from Eqs. (5) and (6) that we must have $\Delta' \parallel \Delta''$ or $\Delta' \perp \Delta''$. The first case corresponds to a sinusoidal and the second to a helicoidal SDW which was considered in Ref. 10.

In the weak coupling model the ground state is, as one can easily verify, a sinusoidal SDW.

The gap in the electron spectrum is determined by the quantity $\text{Det}\Delta^{+-}$. For a sinusoidal SDW the gap is of the same order of magnitude as the order parameter. In the case of a helicoidal wave $\text{Det}\Delta^{+-} = \Delta'^2 - \Delta^2$. The value of this quantity is determined by the λ_{ik} tensor and in the isotropic case $\text{Det}\Delta^{+-} = 0$. For a helicon the gap in the spectrum is thus small compared with the order parameter in as far as the anisotropy energy is small. From measurements of the conductivity in the compounds $(\text{TMTSF})_2\text{AsF}_6$ and $(\text{TMTTF})_2\text{Br}$ it follows that the number of free carriers in turn decreases fast below the antiferromagnetic ordering temperature. The ground state is clearly dielectric. Hence one may conclude that the spin density wave in these compounds is sinusoidal.

The easy z -, the intermediate y -, and the difficult x -magnetization axes are in the model considered the principal axes of the λ_{ik} tensor. The anisotropy tensor is determined apart from an additive term proportional to λ_{ik} which enables us to put the eigenvalue $\lambda_z = 0$. In that case $\lambda_x < \lambda_y < 0$. It turns out that the $(\text{TMTSF})_2\text{AsF}_6$ and $(\text{TMTTF})_2\text{Br}$ salts the principal axes of the anisotropy tensor are the same as the axes of the conductivity tensor. The easy magnetization axis is in this case the b^* axis. The intermediate axis is in the first compound close to a and in the second to c^* [the (a, b^*, c^*) system of the principal axes of the conductivity tensor differs slightly from the triclinic system realized in Bechgaard salts].

In the nesting model⁶ the solution of Eqs. (5) and (6) which is thermodynamically suitable corresponds to the vector $\mathbf{Q}_0 = (2p_F, \pi/b^*, \pi/c^*)$ which guarantees the exact overlap of parts of the Fermi surface for the spectrum (1). It was noted by Gor'kov in his review article¹¹ that the measurement of the Hall effect in the compounds considered indicates the usefulness of the "straight" nesting vector $\mathbf{Q}_1 = (2p_F, 0, 0)$ which must be considered to be an alternative possibility. How one can determine the choice between \mathbf{Q}_0 and \mathbf{Q}_1 is still far from clear. In the framework of the model considered this choice may be connected with the way the kernel of the interaction (3) depends on the momentum transfer in the transverse direction [$\Lambda(\mathbf{Q}_0) \neq \Lambda(\mathbf{Q}_1)$].

Because of the peculiarities of the structure of the Bechgaard salts the ratio t_c/t_b is small⁵ in them ($\sim 1/10-1/30$) so that we neglect in what follows the dependence of the transverse dispersion of the electron spectrum on the component P_c , putting $t_\perp(\mathbf{p}_\perp) = 2t_b \cos \mathbf{p} \cdot \mathbf{b}^*$. The expression for the electron energy takes the form

$$\xi^{(\pm)}(\mathbf{p}) = \pm v_F(p_{\parallel} \mp p_F) + 2t_b \cos \mathbf{p} \cdot \mathbf{b}^*. \quad (7)$$

In the case of straightforward nesting the existence of anomalous averages might lead to the formation of electron-hole "pockets." For this to happen we must have $|\Delta| < 2 \max |t_\perp(\mathbf{p}_\perp)|$. It was shown in Ref. 12 that at $T=0$ for a spectrum of the form (7) the dependence of the order parameter on the transverse dispersion has a "step" form: $|\Delta| = \Delta_0$ when $\Delta_0 > 2t_b$, $\Delta = 0$ when $\Delta_0 < 2t_b$. In that case there are in the excitation spectrum in the magnetic phase at $T=0$ no electron-hole pockets which is confirmed by the disappearance of free carriers at sufficiently low temperatures.

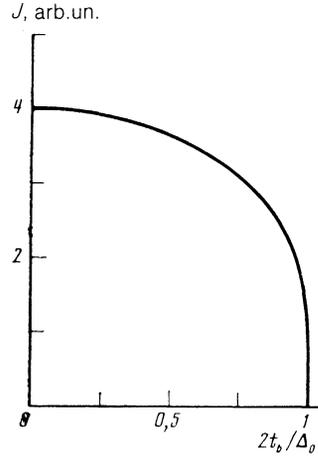


FIG. 1. Transition temperature as function of t_b .

Substituting the solution of the set (5) for $\mathbf{Q} = \mathbf{Q}_1$ into Eq. (6) and changing to integration over $\eta = v_F(p_{\parallel} - p_F)$ we get

$$\frac{1}{\zeta} = \frac{1}{\pi} \int_0^{\pi \omega_0} \int_0^{\pi} \frac{1}{\varepsilon} \text{th} \frac{\varepsilon + 2t_b \cos \varphi}{2T} d\eta d\varphi, \quad (8)$$

where $\zeta = \Lambda/\pi b^* c^* v_F$ and $\varepsilon_2 = \eta^2 + |\Delta|^2$. The frequency ω_0 to cut off the integral is of the order of the Fermi frequency. Equation (8) determines the temperature-dependence in the case of straight nesting. Putting $|\Delta| = 0$ in (8) and integrating over φ we get an equation for the t_b -dependence of the transition temperature:

$$\frac{1}{\zeta} = \int_{2t_b}^{\omega_0} \frac{d\eta}{(\eta^2 - 4t_b^2)^{1/2}} \text{th} \frac{\eta}{2T_c}. \quad (9)$$

The result of a numerical solution of Eq. (9) is shown in Fig. 1. For small T_c

$$\Delta_0 - 2t_b = (2\pi T_c \Delta_0)^{1/2} \exp(-\Delta_0/T_c) \quad (10)$$

(this equation was obtained in Ref. 12 with logarithmic accuracy). When $\Delta_0 = 2t_b$ the transition temperature vanishes. The temperature-dependence of the order parameter for some values of t_b is shown in Fig. 2.

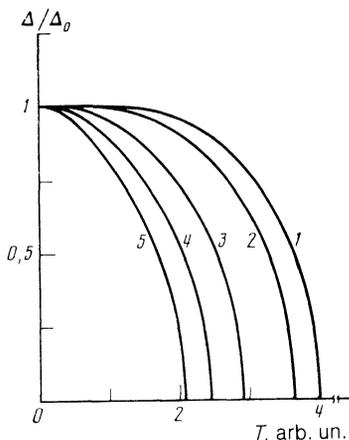


FIG. 2. Temperature-dependence of the order parameter for different values of t_b : 1: $2t_b/\Delta_0 = 0$, 2: 0.5; 3: 0.8; 4: 0.9; 5: 0.95.

3. MAGNETIC PROPERTIES

We use perturbation theory for the evaluation of the spin susceptibility to expand Eq. (5) up to terms of first order in the magnetic field. We need then take into account only the spin part of the interaction. Using the fact that $\Delta_{\alpha\beta}^+ - \Delta_{\beta\gamma}^- = |\Delta|^2 \delta_{\alpha\gamma}$ we get for the electron Green function, say, to the right near the Fermi surface the expression

$$G^{++}(iz, \mathbf{p}) = G_{H=0}^{++}(iz, \mathbf{p}) + \mu_B \frac{(iz - \xi_-)^2 (\sigma \mathbf{H}) + \Delta^{+-} (\sigma \mathbf{H}) \Delta^{-+}}{[(iz - \xi_-)(iz - \xi_+) - |\Delta|^2]^2}, \quad (11)$$

where

$$\begin{aligned} \xi_{\pm} &= \xi(\mathbf{p}) = v_F(p_{\parallel} - p_F) + 2t_{\perp}(p_{\perp}), \\ \xi_{\pm} &= \xi(\mathbf{p} - \mathbf{Q}) = -v_F(p_{\parallel} - p_F) + 2t_{\perp}(p_{\perp} - \mathbf{Q}_{\perp}). \end{aligned}$$

The contribution to the magnetization is given by the formula

$$\mathbf{M} = -\mu_B \text{Sp} [\sigma(G^{++}(0+) + G^{--}(0+))]. \quad (12)$$

Substituting (11) into (12) and writing $\mathbf{n} = \Delta/\Delta$ we get

$$\mathbf{M} = \chi_{\perp}(\mathbf{H} - (\mathbf{Hn})\mathbf{n}) + \chi_{\parallel}(\mathbf{Hn})\mathbf{n}, \quad (13)$$

where

$$\chi_{\perp} = 4\mu_B^2 \frac{T}{(2\pi)^3} \sum_{\mathbf{z}} \int \frac{|\Delta|^2 - (iz - \xi_-)^2}{((iz - \xi_+)(iz - \xi_-) - |\Delta|^2)^2} d^3p, \quad (14a)$$

$$\chi_{\parallel} = 4\mu_B^2 \frac{T}{(2\pi)^3} \sum_{\mathbf{z}} \int \frac{-|\Delta|^2 - (iz - \xi_-)^2}{((iz - \xi_+)(iz - \xi_-) - |\Delta|^2)^2} d^3p. \quad (14b)$$

In what follows all formulae for the susceptibility pertain to the straight nesting case. Using the residue theorem and integrating by parts in the expression for χ_{\perp} we find

$$\chi_{\perp} = 4\mu_B^2 \frac{1}{2\pi b^2 c^* v_F}, \quad (15a)$$

$$\begin{aligned} \chi_{\parallel} &= 2\mu_B^2 \frac{1}{(2\pi)^3 v_F} \int_0^{\infty} \left[\left(2T \text{ch}^2 \frac{\varepsilon + 2t_{\perp}(p_{\perp})}{2T} \right)^{-1} \right. \\ &\quad \left. + \left(2T \text{ch}^2 \frac{\varepsilon - 2t_{\perp}(p_{\perp})}{2T} \right)^{-1} \right] d^2p_{\perp} d\eta \end{aligned} \quad (15b)$$

(the equivalent expression for the case of ideal nesting was obtained in Ref. 13).

The transverse susceptibility in the model considered is thus identically equal to the spin susceptibility of a normal metal $\chi_n = \mu_B^2 N(0)$ where $N(0) = 2/\pi b^2 c^* v_F$ is the density of states at the Fermi surface. As for the dispersion law (7) the formation of electron-hole pockets is impossible at zero temperature the longitudinal susceptibility vanishes at $T = 0$. This result is in agreement with the experimental data for³ (TMTSF)₂AsF₆ and for⁴ (TMTTF)₂Br.

Near the transition point the longitudinal susceptibility changes linearly with temperature. The derivative of the longitudinal susceptibility for $|\Delta| \ll T_c$ is

$$\frac{\partial \chi_{\parallel}}{\partial T} = \chi_n \frac{2}{T_c} \int_{2t_b}^{\infty} \frac{\eta d\eta}{(\eta^2 - 4t_b^2)^{1/2} \cdot 2T \text{ch}^2 \eta / 2T}. \quad (16)$$

For $T_c \ll \Delta_0$ the slope becomes small:

$$\frac{\partial \chi_{\parallel}}{\partial T} = \chi_n \frac{2}{T_c} \left(\frac{2\pi \Delta_0}{T_c} \right)^{1/2} \exp\left(-\frac{\Delta_0}{T_c} \right). \quad (17)$$

Using (10) we can rewrite this expression in the form

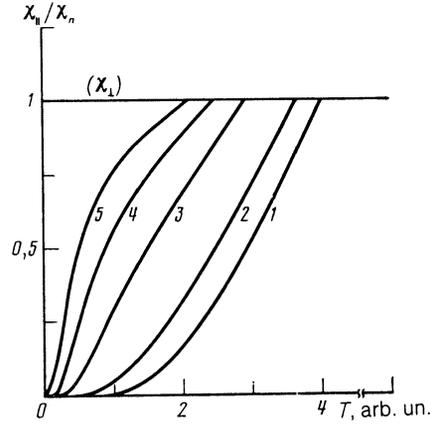


FIG. 3. Temperature-dependence of the spin susceptibility for different values of t_b : 1: $2t_b/\Delta_0 = 0$; 2: 0.5; 3: 0.8; 4: 0.9; 5: 0.95.

$$\frac{\partial \chi_{\parallel}}{\partial T} = 2\chi_n \frac{\Delta_0 - 2t_b}{T_c^2}. \quad (18)$$

For $t_b = 0$ Eq. (16) gives $\partial \chi_{\parallel} / \partial T = 2\chi_n / T_c$. We show in Fig. 3 the result of a numerical calculation of the temperature dependence of the longitudinal spin susceptibility. The dependence obtained agrees qualitatively with experiments for (TMTSG)₂AsF₆ (Ref. 3) and (TMTTF)₂Br (Ref. 4). We discuss below the problem of a quantitative agreement.

We turn to the evaluation of the spin-flop field. As we noted above the easy, intermediate, and difficult magnetization axes are the principal z, y, x axes of the anisotropy tensor. The easy axis $z \parallel \mathbf{b}^*$. The spin rotation occurs in the yz plane. When $H > H_{sf}$ the magnetization of the sublattices is parallel to the intermediate axis.

Near T_c the change in the order parameter when the field is switched on is $\mu_B^2 H^2 / |\Delta|$. We shall assume in what follows that $(\mu_B H / T_c)^2 \ll (T_c - T) / T_c$. We can then use the thermodynamic identity which connects the derivatives of the Hamiltonian and the thermodynamic potential with respect to the parameter:

$$\delta \Omega / \delta \xi = \langle \partial \hat{H} / \partial \xi \rangle_{gr.st.} \quad (19)$$

Formally differentiating the Hamiltonian with respect to μ_B we get for the change in the thermodynamic potential when the field is switched on the obvious expression

$$\delta \Omega_1 = -1/2 \delta \mathbf{M} \mathbf{H}. \quad (20)$$

We introduce the angle θ between the direction of the magnetic field which is parallel to the easy axis and the vector Δ . Substituting (13) into (20) we get

$$\delta \Omega_1 = -1/2 H^2 (\chi_{\perp} - \chi_{\parallel}) \delta \sin^2 \theta. \quad (21)$$

The term in the Hamiltonian which describes the anisotropic part of the effective interaction has the form

$$V_{eff} = 1/4 \lambda_{ik} \sigma_{\alpha\gamma} \sigma_{\beta\delta} \psi_{\alpha}^+ (\mathbf{p}_+) \psi_{\beta}^+ (\mathbf{p}_+) \psi_{\gamma} (\mathbf{p}_-) \psi_{\delta} (\mathbf{p}_-). \quad (22)$$

The anisotropy is small which allows us to use again Eq. (19). The response of the thermodynamic potential to a rotation of the sublattice magnetization is

$$\delta \Omega_2 = -2 \frac{|\Delta|^2}{\Lambda^2} \lambda_{\nu} \delta \sin^2 \theta. \quad (23)$$

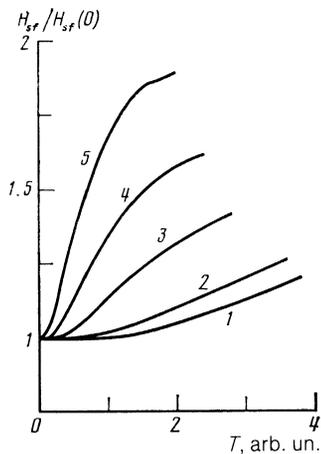


FIG. 4. Temperature dependence of the spin-flop field for different values of t_b : 1: $2t_b/\Delta_0 = 0$; 2: 0.5; 3: 0.8; 4: 0.9; 5: 0.95.

The total change in the potential is $\delta\Omega = \delta\Omega_1 + \delta\Omega_2$. The equilibrium value of the angle θ minimizes the quantity $\delta\Omega$. One sees easily that the spin flop is a first order transition and

$$H_{sf} = \left(\frac{4|\Delta|^2(-\lambda_y)}{\Lambda^2(\chi_\perp - \chi_\parallel)} \right)^{1/2}. \quad (24)$$

Since $\chi_\parallel = 0$ but $\chi_\perp = \chi_n$ at $T = 0$ for the spectrum (7)

$$H_{sf}(0) = \frac{1}{\mu_B} \left(\frac{4|\Delta|^2(-\lambda_y)}{\Lambda^2} \times \frac{\pi b^* c^* v_F}{2} \right)^{1/2}. \quad (25)$$

One can obtain a formula which explicitly determines the temperature dependence of H_{sf} by substituting Eqs. (15a), (15b) in (24) (Fig. 4). In the case of ideal overlapping of parts of the Fermi surface the values of the spin-flop field at zero temperature and near the transition point are connected by the relation

$$H_{sf}(T_c)/H_{sf}(0) = 2\gamma/(7\zeta(3))^{1/2} \approx 1.23. \quad (26)$$

If the superstructure vector is straight, $H_{sf}(T_c)$ increase with increasing t_b becoming infinite at $t_b = \Delta_0/2$ (Fig. 5).

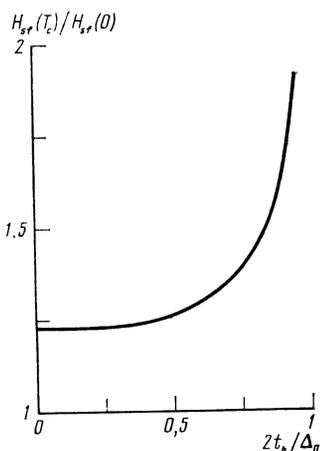


FIG. 5. H_{sf} as function of t_b close to the transition point.

To evaluate the antiferromagnetic resonance frequency we apply the method of analytical continuation of thermodynamic responses expounded in Ref. 14. The quantity ω_{af} is determined from the self-consistency equation for a small addition to the order parameter. Detailed calculations are given in the Appendix. At $T = 0$ the expression for the antiferromagnetic resonance frequency has the form

$$\omega_{af}^{+(-)} = 2 \left(\frac{4|\Delta|^2(-\lambda_{x(y)})}{\Lambda^2} \frac{\pi b^* c^* v_F}{2} \right)^{1/2}. \quad (27)$$

The + and - indices refer to the polarization of oscillations which are, respectively, parallel to the difficult and the intermediate magnetization axes. As $\lambda_x < \lambda_y < 0$, we have $\omega_{af}^+ > \omega_{af}^-$.

One sees easily that as there are no electron-hole pockets at $T = 0$ we have the relation $\omega_{af}^+ = 2\mu_B H_{sf}$ which is also valid in the localized spin theory. Experimental data for $(\text{TMTSF})_2\text{AsF}_6$ (Refs. 2 and 3) and $(\text{TMTTF})_2\text{Br}$ (Ref. 4) indicate that the relation considered is, indeed, satisfied in these compounds at sufficiently low temperatures.

4. CONCLUSION

The results obtained thus indicate the applicability of a model with two open parts of the Fermi surface for a qualitative description of the magnetic properties of the $(\text{TMTSF})_2\text{AsF}_6$ and $(\text{TMTTF})_2\text{Br}$ compounds. In this model an antiferromagnetic has, just as in the model with localized spins, an anisotropic spin susceptibility, a spin-flop field, and an antiferromagnetic-resonance frequency.

It turns out that at zero temperature the predictions of the delocalized model with the spectrum (7) is independent of the choice of the superstructure vector. The ground state in the compounds considered is at $T = 0$ apparently dielectric which confirms the conclusion that at zero temperature there are no electron-hole pockets in the quasi-particle spectrum. The relation $\omega_{af}^- = 2\mu_B H_{sf}$ obtained for $T = 0$ is to a good accuracy checked by measurements in the compounds $(\text{TMTSF})_2\text{AsF}_6$ and $(\text{TMTTF})_2\text{Br}$ at sufficiently low temperatures. One must note that the relation considered is, generally speaking, violated in the case when electron-hole pockets are formed, if the transverse dispersion law allows their existence.

At finite temperatures the theoretical curve of the temperature dependence of the spin susceptibility agrees only qualitatively with experiment. In that case it is for $(\text{TMTSF})_2\text{AsF}_6$ closest to the experimental curve corresponding to ideal overlap. On the other hand, in the compound $(\text{TMTTF})_2\text{Br}$ the behavior of the $\chi_\parallel(T)$ curve indicates a difference between the superstructure vector and the vector \mathbf{Q}_0 realizing ideal overlap.

In the model considered the ratio of the values of the spin flop field at T_c and at zero temperature is always larger than unity. The minimum value $H_{sf}(T_c)/H_{sf}(0) \approx 1.23$ is reached for ideal nesting. Experimentally, H_{sf} in $(\text{TMTSF})_2\text{AsF}_6$ is practically independent of the temperature which again indicates that in that compound the superstructure vector is close to \mathbf{Q}_0 . Unfortunately, in $(\text{TMTTF})_2\text{Br}$ the quantity H_{sf} has been measured⁴ only at $T = 4.2$ K, but the fact that at that temperature the relation $\omega_{af}^- = 2\mu_B H_{sf}$ is satisfied enables us to conclude that the difference between the overlap vector and \mathbf{Q}_0 can at any rate not be too large.

The "Shubnikov-de Haas" oscillation frequencies in $(\text{TMTSF})_2\text{PF}_6$ observed in Ref. 15 turned out to correspond to a series of exciton transitions in the magnetic field. As the areas of the orbits are in that case not too small ($\sim 1\%$ of the Brillouin zone cross section) the conclusion was reached in Ref. 15 that in Bechgaard salts there exists an antiferromagnetic structure with a straight overlap vector. Comparing this statement with the results obtained above we are led to the conclusion that the SDW vector may depend on the magnetic field.

The author expresses his deep gratitude to L. P. Gor'kov for suggesting the subject and for supervision of this work.

APPENDIX

Calculation of the antiferromagnetic resonance frequency

We write Eq. (5) in matrix form. The perturbation of the Hamiltonian is

$$\hat{h}_\omega^{(1)} = \begin{pmatrix} 0 & \Delta_\omega^{(1)+-} \\ \Delta_\omega^{(1)-+} & 0 \end{pmatrix}. \quad (\text{A1})$$

The response in the matrix Green function is

$$\hat{G}_{z,z-\omega}^{(1)} = -\hat{G}_z^{(0)} \hat{h}_\omega^{(1)} \hat{G}_{z-\omega}^{(0)}. \quad (\text{A2})$$

Substituting in this expression the solution of the set (5) when there is no external field we get

$$G_{z,z-\omega}^{(1)+-}(\mathbf{p}) = \frac{[z-2t_\perp(\mathbf{p}_\perp) + \eta][z-2t_\perp(\mathbf{p}_\perp) - \omega - \eta] \Delta_\omega^{(1)+-} + \Delta^{+-} \Delta_\omega^{(1)-+} \Delta^{+-}}{[(z-2t_\perp(\mathbf{p}_\perp))^2 - \varepsilon^2][(z-2t_\perp(\mathbf{p}_\perp) - \omega)^2 - \varepsilon^2]} \quad (\text{A3})$$

The self-consistency equation for the perturbation as the form

$$\Delta_\omega^{(1)+-} = (\Lambda\delta_{ik} + \lambda_{ik}) \sigma_i \frac{1}{2} \text{Sp} \left[\frac{T}{(2\pi)^3} \sum_z \int \sigma_k G_{z,z-\omega}^{(1)+-}(\mathbf{p}) d^3p \right]. \quad (\text{A4})$$

One sees easily that the equations have a solution when $\Delta^{(1)} \perp \Delta$, i.e., in the commensurate case the oscillations are transverse. To evaluate the sum over the frequencies the thermodynamic functions must be continued onto the real axis. The analytic continuation is realized by the formula

$$T \sum_z G_{z,z-\omega}^{(1)} = - \int_{-\infty}^{+\infty} \frac{dz}{4\pi i} \left\{ \text{th} \frac{z-\omega}{2T} \hat{G}_z^{(0)R} \hat{h}_\omega^{(1)} \hat{G}_{z-\omega}^{(0)R} - \text{th} \frac{z}{2T} \hat{G}_z^{(0)A} \hat{h}_\omega^{(1)} \hat{G}_{z-\omega}^{(0)A} + \left(\text{th} \frac{z}{2T} - \text{th} \frac{z-\omega}{2T} \right) \hat{G}_z^{(0)R} \hat{h}_\omega^{(1)} \hat{G}_{z-\omega}^{(0)A} \right\}. \quad (\text{A5})$$

The fact that for the spectrum (7) $t_\perp(\mathbf{p}_\perp) = -t_\perp(\mathbf{p}_\perp + \mathbf{Q}_\perp)$ enables us to simplify the writing down of the formulae. If we neglect the scattering of free carriers by phonons and impurity atoms the evaluation of the integral gives ($\omega \ll T$)

$$T \sum_z G_{z,z-\omega}^{(1)+-}(\mathbf{p}) = \Delta_\omega^{(1)+-} \frac{1}{2\varepsilon} \text{th} \frac{\varepsilon + 2t_\perp(\mathbf{p}_\perp)}{2T} + \Delta_\omega^{(1)+-} \frac{\omega^2}{8} \frac{1}{\varepsilon(\varepsilon^2 - \omega^2/4)} \times \text{th} \frac{\varepsilon + 2t_\perp(\mathbf{p}_\perp)}{2T}. \quad (\text{A6})$$

Substituting this expression into Eq. (A4) we find

$$-\frac{\lambda_{x(y)}}{\Lambda} = \frac{\omega^2}{4} \frac{\Lambda}{(2\pi)^3 v_F} \int \int \text{th} \frac{\varepsilon + 2t_\perp(\mathbf{p}_\perp)}{2T} \frac{d\eta d^2 p_\perp}{\varepsilon(\varepsilon^2 - \omega^2/4)}. \quad (\text{A7})$$

Equation (A7) plays the role of a dispersion relation for ω_{af} . Evaluating the integral for $T = 0$ and using the fact that there are no electron-hole pockets at zero temperature we get Eq. (27). Near the transition point $|\Delta| \ll T$ and the integral in (A7) is easily performed. One should note that in that temperature range ω_{af} is independent of the form of the function $t_\perp(\mathbf{p}_\perp)$. When $\omega > 2\delta$ there is an imaginary term in the reso-

nance frequency corresponding to an absorption of the oscillations. If $\omega \ll |\Delta| \ll T$ we have

$$\omega_{af}^{+(-)} = \left(\frac{64|\Delta|T(-\lambda_{x(y)})}{\Lambda^2 \pi} \frac{\pi b^* c^* v_F}{2} \right)^{1/2}. \quad (\text{A8})$$

If $|\Delta| \ll |\omega| \ll T$ the main contribution to the integral comes from the vicinity of the pole of the integrand. In that limiting case the resonance frequency becomes purely imaginary which corresponds to the diffusive mode. We now write down the dispersion relation taking relaxation processes into account. For the sake of simplicity we put $\omega \ll |\Delta|$. In the case of isotropic scattering one can get the result introducing an imaginary term in the frequency of the Green functions in (A5): $z \rightarrow z \pm i/2\tau$. We have

$$-\frac{\lambda_{x(y)}}{\Lambda} = \frac{\omega^2}{4} \frac{\Lambda}{(2\pi)^3 v_F} \int \int \text{th} \frac{\varepsilon + 2t_\perp(\mathbf{p}_\perp)}{2T} \frac{d\eta d^2 p_\perp}{\varepsilon^3} + i \frac{\omega \Lambda}{4\tau (2\pi)^3 v_F} \int \int \frac{d\eta d^2 p_\perp}{2T \varepsilon^2 \text{ch}^2[(\varepsilon + 2t_\perp(\mathbf{p}_\perp))/2T]}. \quad (\text{A9})$$

If $T_c - T \ll T_c$ the dispersion relation takes the form

$$\omega^2 + i \frac{\omega}{\tau} - \frac{64|\Delta|T(-\lambda_{x(y)})}{\Lambda^2 \pi} \frac{\pi b^* c^* v_F}{2} = 0. \quad (\text{A10})$$

When $T = 0$ the evaluation of the integrals in (A9) gives the earlier result (27) for ω_{af} . As one should expect when there are no electron-hole pockets the damping vanishes.

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