Pauli coupling of the external magnetic field to spin-density waves

A. Bjeliš and D. Zanchi

Department of Physics, Faculty of Science, University of Zagreb, P.O. Box 162, 41001 Zagreb, Croatia

The effects of the external magnetic field on the spin-density-wave (SDW) order and on accompanying fluctuations are calculated within the random-phase approximation for the extended Hubbard model with imperfectly nested quasi-one-dimenisonal Fermi surfaces. Both Pauli and orbital mechanisms are treated in parallel. It is shown that the Pauli coupling leads to a finite hybridization of the SDW component in the direction of the external field and the charge-density wave. The mean value of this SDW component remains zero below the critical temperature in the isotropic system, but may be activated in systems with an internal spin anisotropy. The mean-field expression for the corresponding spin-flop field is derived. Furthermore, the Pauli coupling renormalizes two of six fluctuative SDW modes. In order to establish ways of qualitative and quantitative comparison between effects belonging to the Pauli and orbital couplings, we analyze the characteristic parameters for these two modes as well as for the other four modes affected only by the orbital coupling. In particular we evaluate the field dependence of long-wavelength gaps, correlation lengths, and attenuation coefficients.

I. INTRODUCTION

The electronic spin-density waves (SDW's) are by now found and studied mostly in quasi-one-dimensional Bechgaard salts $(TMTSF)_2X$. The characteristic temperatures of three-dimensional SDW order in these materials are rather low (~ 10 K). In this temperature range external magnetic fields of moderate and large strengths ($H < 20$ T) act as a nontrivial (i.e., nonlinear) probe which may alter dramatically the properties of the order itself and of the critical fluctuations as well. The most significant phenomena like a cascade of field-induced SDW (FISDW) states,[1-4] resonances in magnetoresistance and magnetic torque at particular orientations of magnetic field,[5,6] and anomalous increase of, e.g., magnetoresistivity[7,8] are all interpreted as consequences of the orbital coupling to the magnetic field. In the present work we consider both the orbital and Pauli couplings of the external magnetic field to the band electrons. Our aim is to point out the effects of the latter on the SDW properties, and to compare them with the well-known effects of the orbital coupling.

The finite orbital coupling goes together with the creation of pockets at the Fermi surface consisting of two folded quasi-one-dimensional sheets in the SDW state. It was claimed that the pockets due to the imperfect nesting (i.e., finite effective next-neighboring transverse hopping ε_0) are responsible for FISDW's (Refs. 9–11) and Lebeds' resonances.[12,13] (For simplicity we consider only the b transverse direction.) Even if the nesting is perfect ($\varepsilon_0 = 0$ and direct transverse hopping t_b finite), the orbital coupling induced by spatial fluctuations causes the decrease of the transverse correlation length ξ_b,[14] with the mentioned effects on the resistivity, specific heat, etc. In any case the characteristic inverse magnetic length and the energy associated to the orbital coupling are $q_0 = ebH \cos\theta$ and $\omega_0 = v_f q_0$, respectively. Here θ is the inclination of the magnetic field $\mathbf{H}$ from the transverse c

direction in the (b,c) plane. On the other hand, the corresponding scales for the Pauli coupling are $q_P = \mu_B H / v_F$ and $\omega_P = \mu_B H$. The ratio of two scales is

$$\eta^{-1} \equiv q_P / q_0 = \omega_P / \omega_0$$

$$= \mu_B /(ebv_F \cos\theta) = 2am_{\text{eff}} \cot(k_F a)/bm \cos\theta ,$$

where m is the bare electron mass and m_{eff} is the effective mass corresponding to the longitudinal dispersion,

$$m_{\text{eff}} = (2t_a a^2 \cos k_F a)^{-1} = \tan(k_F a)/v_F a .$$

Being a product of competing geometric ($a/b < 1$) and electronic ($m/m_{\text{eff}} > 1$) factors, this ratio has in real materials values which do not discriminate strongly any of two mechanisms. In other words, one expects comparable contributions from orbital and Pauli coupling to those properties which are sensitive to both of them. In order to specify the properties of this kind, it is worthwhile to evoke some basic facts regarding the symmetry of SDW in the magnetic field.

The orbital coupling does not affect the degeneracy of spin orientation (or its preferable direction due to the internal spin anisotropy). In a standard approach one fixes the direction of spin in the rectilinear state and looks for the orbital effects in the parameters of the Landau expansion for the phason propagator and the free energy. The treatments of SDW's and CDW's are then completely equivalent. The Pauli coupling lifts the spin degeneracy, enforcing the spin orientation in the plane perpendicular to $\mathbf{H}$. As will be shown below, this orthogonality is realized irrespectively to the details of electron spectrum, interactions etc. Then the Zeeman splitting has no influence on the intraplanar SDW properties, but it affects the spin fluctuations out of plane, i.e., two of six collective SDW modes. In the present work we calculate the parameters characterizing these two modes in the

temperature range above and below T_c, taking into account both Pauli and orbital couplings.

The Pauli coupling involves also the CDW component. In fact, there is a finite off-diagonal term in the total $2k_F$ susceptibility, i.e., the Zeeman splitting of spin-up and spin-down components leads to a finite SDW-CDW coupling. The corresponding eigenmodes are thus hybridized SDW-CDW fluctuations. This hybridization is even more important in the CDW systems, since it causes the dependence of the critical temperature and the wave vector of the ordering on $\mathbf{H}$.[15,16]

Having in mind the above remarks we start in Sec. II from the extended Hubbard model and derive the total 4×4 matrix response for the $2k_F$ density waves within the random-phase approximation (RPA), with orbital and Pauli terms fully (i.e., nonperturbatively) taken into account. The SDW-CDW hybridization and the combined

influence of external magnetic field and intrinsic spin anisotropy on the SDW ordering are discussed in Sec. III. The analysis of the collective modes at $T > T_c$ and $T < T_c$ are given in Sec. IV. Section V contains concluding remarks.

II. DENSITY-WAVE SUSCEPTIBILITY MATRIX

A. Hamiltonian

We start from the usual tight-binding model for the anisotropic two-dimensional band with the Fermi surface which is open in the transverse ($\hat{b}$) direction. The longitudinal ($\hat{a}$) axis corresponds to the best conductivity (i.e., chain) direction in Bechgard salts. In the absence of external magnetic field the kinetic part of the Hamiltonian can be written as

$$H_{\text{band}} = \frac{b}{4\pi} \int dp_2 \int dx \, \Psi^\dagger(x,p_2)[iv_F\rho_3\partial_x - 2t_b\cos(p_{2b}) - \varepsilon_0\cos(2p_2b)]\Psi(x,p_2) \ . \tag{1}$$

Here $\Psi^\dagger$ and Ψ are four-component fermion fields,

$$\Psi^\dagger = (\Psi^\dagger_{\uparrow+}, \Psi^\dagger_{\uparrow-}, \Psi^\dagger_{\downarrow+}, \Psi^\dagger_{\downarrow-})$$

with the second index $+$ ($-$) denoting the right (left) sheet of the Fermi surface. ρ_i's are the Pauli matrices in this two-dimensional space. Indices $\uparrow, \downarrow$ span the spin space. The longitudinal dispersion in (1) is linearized in the vicinity of the Fermi wave numbers $\pm k_F$, with v_F being the Fermi velocity in the x direction. The ε_0 term is the quadratic correction to this linearization,[17] transposed into the transverse dispersion as an effective second-neighbor hopping.[18] It parametrizes the imperfect nesting. The t_b term represents the real hopping between nearest-neighboring chains. Finally $2\mathbf{Q} = (2Q_1, 2Q_2) = (2k_F, \pi/b)$ is the nesting wave vector for the spectrum (1) with $\varepsilon_0 = 0$.

Let us now introduce the uniform external magnetic field perpendicular to the chain direction. The corresponding vector potential can be chosen as

$$\mathbf{A}(\mathbf{r}) = H(\mathbf{j}\cos\theta - \mathbf{k}\sin\theta)x \ ,$$

where $\mathbf{j}$ and $\mathbf{k}$ are unit vectors along $\hat{b}$ and $\hat{c}$ axes, respectively. The orbital coupling of the magnetic field to the band electrons enters through the standard Peierls substitution, and the Pauli paramagnetic term is introduced into H_{band} by putting the spin quantization (z) axis in the direction of $\mathbf{H}$. Thus, the band in the magnetic field is represented by

$$H_0 = \frac{b}{4\pi} \int dp_2 \int dx \, \Psi^\dagger(x,p_2)[H_{1D} + H_{Q1D,orb}$$

$$+ H_{\text{Pauli}}]\Psi(x,p_2) \tag{2}$$

with

$$H_{1D} = iv_F\rho_3\partial_x \ , \tag{3a}$$

$$H_{Q1D,orb} = -2t_b\cos(p_2 + \rho_3Q_2 - A_2)b$$

$$-\varepsilon_0\cos2(p_2 + \rho_3Q_2 - A_2)b \ , \tag{3b}$$

$$H_{\text{Pauli}} = -\sigma_3\mu_B H \ . \tag{3c}$$

μ_B is the Bohr magneton, and we put $e = c = 1$ and $A_2 \equiv hx\cos\theta$ into Eq. (3b).

The quasi-one-dimensional Hamiltonian (2) with no manifest translational invariance in the x direction can be still reduced to a pure one-dimensional effective Hamiltonian by the procedure proposed by Gor'kov and Lebed.[9] One introduces a new fermion field

$$\Psi(x,p_2) = \exp[-i\hat{\Phi}(x,p_2)]\bar{\Psi}(x,p_2) \tag{4}$$

with the phase operator $\hat{\Phi}$ given by

$$\hat{\Phi}(x,p_2) = v_F^{-1}\rho_3 \int^x dx \, H_{Q1D,orb}(x,p_2) \ . \tag{4'}$$

Represented in terms of the fields $\bar{\Psi}$ and $\bar{\Psi}^\dagger$ the Hamiltonian H_0 has the form (2) but with the term $H_{Q1D,orb}$ absent. On the other hand, we could reduce the Hamiltonian (2) to that containing only $H_{1D} + H_{Q1D,orb}$ by performing the transformation

$$\Psi(x,p_2) = \exp[i\hat{\chi}(x)]\hat{\Psi}(x,p_2) \ , \tag{5}$$

where the phase operator $\hat{\chi}(x)$ reads

$$\hat{\chi}(x) = -v_F^{-1}\rho_3 \int^x dx \, H_{\text{Pauli}} = v_F^{-1}\rho_3\sigma_z\mu_B Hx \ . \tag{5'}$$

Applying the transformation (5) to the field $\hat{\Psi}$, one introduces finally the field $\tilde{\Psi}$ for which the corresponding Hamiltonian H_0 is purely one-dimensional, i.e., contains only the term H_{1D}. Thus, by performing successively transformation (4) and (5) one passes from the problem of diagonalization of the Hamiltonian (2) (completed by the interaction part) to the determination of the eigenstates

and propagators dressed by the phase factors (4') and (5').

The dominant interaction responsible for the SDW order in Bechgaard salts is Coulomb interaction, in particular, its short-range part. The corresponding contributions to the Hamiltonian can be written as

$$H_{\text{int}} = \frac{2\pi v_F}{8} \int dx \sum_{\mathbf{R}\perp} [-g_2 \Psi^\dagger \sigma_i \rho_- \Psi \Psi^\dagger \sigma_i \rho_+ \Psi$$

$$+ (2g_1 - g_2) \Psi^\dagger \rho_- \Psi \Psi^\dagger \rho_+ \Psi$$

$$+ (\text{same with } \rho_- \leftrightarrow \rho_+)] , \qquad (6)$$

with g_1 and g_2 being backward and forward coupling constants, respectively, and $\rho_\pm \equiv \rho_1 \pm i\rho_2$. Here we omit the Umklapp terms and contributions in which all four electronic states are on the same (left or right) Fermi sheet. In the particular case of a pure Hubbard model with the on-site terms only, one has $g_1 = g_2 \equiv U/2\pi v_F$. From now on we use the notation $2\pi v_F g_2 \equiv U_s$, $2\pi v_F(2g_1 - g_2) \equiv U_c$ and allow for the extensions of the Hubbard model which include also the interatomic contributions.[19] Furthermore, we introduce the density-wave operators defined by

$$M_i = \tfrac{1}{2} \Psi^\dagger \rho_+ \sigma_i \Psi , \quad i = 1,2,3,4 , \qquad (7)$$

where $\sigma_4 \equiv I$. The first three components ($i = 1,2,3$) define the complex SDW vector amplitude, while the fourth component $M_4 \equiv C$ is the complex CDW scalar amplitude. Expressed in terms of operators (7), the interaction term (6) reads

$$H_{\text{int}} = \tfrac{1}{2} \int dx \sum_{\mathbf{R}_\perp} [-U_s \mathbf{M}^\dagger(\mathbf{R}) \cdot \mathbf{M}(\mathbf{R}) + U_c C^\dagger(\mathbf{R}) C(\mathbf{R})$$

$$- U_s \mathbf{M}(\mathbf{R}) \cdot \mathbf{M}^\dagger(\mathbf{R}) + U_c C(\mathbf{R}) C^\dagger(\mathbf{R})] . \qquad (8)$$

Note that for the pure Hubbard case the effective SDW and CDW coupling constants are of the equal strengths and opposite signs.

B. RPA susceptibility matrix

The susceptibility matrix for the density waves (DW's) is defined as

$$\chi_{ij}(\mathbf{q}, t - t') = -\theta(t - t') \langle [M_i(\mathbf{q}, t), M_j^\dagger(\mathbf{q}, t')] \rangle$$

$$\equiv \langle M_i M_j^\dagger \rangle , \quad i = 1, \ldots, 4 . \qquad (9)$$

Here it will be derived in two steps. At first, we calculate the Hartree-Fock matrix χ_{ij}^0 for the noninteracting system (1), by using above phase transformations (4) and (5). Then we include interactions (8) and construct in a standard way the Dyson system of equations for the RPA susceptibility.

In order to calculate χ_{ij}^0 it is convenient to make the transformation (5) and pass to the fermion field $\hat{\Psi}$. The DW operators are then given by

$$M_{1,2}\{\Psi\} = M_{1,2}\{\hat{\Psi}\} ,$$

$$M_{3,4}\{\Psi\} = \tfrac{1}{2}(M_3\{\hat{\Psi}\} + M_4\{\hat{\Psi}\})e^{-2iq_p x} \qquad (10)$$

$$\pm \tfrac{1}{2}(M_3\{\hat{\Psi}\} - M_4\{\hat{\Psi}\})e^{2iq_p x} ,$$

where all fermion fields are in the x representation and $M_i\{\hat{\Psi}\}$ are defined by Eq. (7) with the replacement $\Psi \to \hat{\Psi}$. The elements of the matrix χ_{ij}^0 can be now expressed through those of the matrix $\langle M_i\{\hat{M}\} M_j^\dagger\{\hat{\Psi}\} \rangle$. The latter is diagonal since it represents the HF susceptibility for $H_{1D} + H_{Q1D,\text{orb}}$. Its Fourier transform is

$$\langle M_i\{\hat{\Psi}\} M_j^\dagger\{\hat{\Psi}\} \rangle = \delta_{ij} \chi_0(\mathbf{q}, \omega_n) , \qquad (11)$$

where χ_0 is the bare (bubble) susceptibility containing orbital contributions of the magnetic field. The matrix elements of the original HF susceptibility follow immediately from Eqs. (10) and (11). The nonvanishing matrix elements are

$$\chi_{11}^0 = \chi_{22}^0 = \chi^0 ,$$

$$\chi_{33}^0 = \chi_{44}^0 = \tfrac{1}{2}(\chi_\uparrow + \chi_\downarrow) , \qquad (12)$$

$$\chi_{34}^0 = \chi_{43}^0 = \tfrac{1}{2}(\chi_\uparrow - \chi_\downarrow) ,$$

with

$$\chi_{\uparrow,\downarrow}(\mathbf{q}, \omega_n) \equiv \chi_0(q_1 \pm 2q_p, q_2, \omega_n) . \qquad (12')$$

The calculation of χ_0 proceeds via the phase transformation (4) from the field $\hat{\Psi}$ to the one-dimensional field $\tilde{\Psi}$. The corresponding relation between Green's functions is

$$G_{Q1D,\text{orb}}^0(x, x', p_2, \omega_n)$$

$$= \exp\{-i[\hat{\Phi}(x', p_2) - \hat{\Phi}(x, p_2)]\} G_{1D}^0(x - x', \omega_n) \qquad (13)$$

with G_{1D}^0 being the diagonal Green's function for the H_{1D}. χ_0 can be expressed as the 1D bubble "dressed" by the phase factors. After straightforward steps one obtains the expression which has the form of an infinite series

$$\chi_0(\mathbf{q}, \omega_n) = \sum_{l=-\infty}^{\infty} P(k - lq_0, \omega_n) I_l(p_2) \qquad (14)$$

and is manifestly translational invariant. Here $P(k)$ is the 1D bubble

$$P(k, \omega_\nu) = -T \sum_n \frac{1}{2\pi} \int dk' G_{1D,11}^0(k', \omega_n)$$

$$\times G_{1D,33}^0(k' + k, \omega_n + \omega_\nu) , \qquad (15)$$

and the coefficients I_l contain both the dependence on the transverse wave number and orbital effects of magnetic field. They are given by[13,14]

$$I_l(p_2) = \left[\sum_{l'} J_{l-2l'} \left[\frac{4t_b}{v_F q_0} \sin\frac{p_2 b}{2} \right] J_{l'} \left[\frac{2t_b}{v_F q_0} \cos p_2 b \right] \right]^2 . \qquad (16)$$

Once the HF susceptibility matrix χ_{ij}^0 is known, we can construct the Dyson system for the RPA susceptibilities χ_{ij}, with the interactions (8) taken into account. One gets the system of equations

$$\chi_{11,22} = \chi_{11,22}^0 + U_s \chi_{11,22}^0 \chi_{11,22} \; ,$$

$$\chi_{33} = \chi_{33}^0 + U_s \chi_{33}^0 \chi_{33} - U_c \chi_{34}^0 \chi_{43} \; ,$$

$$\chi_{44} = \chi_{44}^0 - U_c \chi_{44}^0 \chi_{44} + U_s \chi_{43}^0 \chi_{34} \; , \qquad (17)$$

$$\chi_{34} = \chi_{34}^0 - U_c \chi_{34}^0 \chi_{44} + U_s \chi_{33}^0 \chi_{34} \; ,$$

$$\chi_{43} = \chi_{43}^0 - U_c \chi_{44}^0 \chi_{43} + U_s \chi_{43}^0 \chi_{33} \; .$$

The solution of this system is

$$[\chi_{ij}(\mathbf{q},\omega)] = \begin{vmatrix} \dfrac{\chi_0(\mathbf{q},\omega)}{1-U_s\chi_0(\mathbf{q},\omega)} & 0 & 0 & 0 \\[3mm] 0 & \dfrac{\chi_0(\mathbf{q},\omega)}{1-U_s\chi_0(\mathbf{q},\omega)} & 0 & 0 \\[3mm] 0 & 0 & \dfrac{\chi_g(\sqrt{1+\delta^2}+U_c\chi_g)}{D} & \chi_g\delta/D \\[3mm] 0 & 0 & \chi_g\delta/D & \dfrac{\chi_g(\sqrt{1+\delta^2}-U_s\chi_g)}{D} \end{vmatrix} \; , \qquad (18)$$

with

$$\chi_g \equiv \sqrt{\chi_\uparrow(\mathbf{q},\omega)\chi_\downarrow(\mathbf{q},\omega)} \; , \qquad (19a)$$

$$\delta \equiv [\chi_\uparrow(\mathbf{q},\omega)-\chi_\downarrow(\mathbf{q},\omega)]/2\chi_g \; , \qquad (19b)$$

$$D \equiv 1+(U_s-U_c)\chi_g\sqrt{1+\delta^2}-U_cU_s\chi_g^2 \; . \qquad (19c)$$

As is seen from Eqs. (17) and (18) the Zeeman splitting induces a mixing of the CDW and the component of SDW parallel to the magnetic field. In Secs. III and IV we shall consider the consequences of this mixing separately at $T>T_c$ and $T<T_c$. The (M_3,M_4) block of χ_{ij} can be diagonalized. The eigenvalues are

$$\chi_\pm = \frac{\chi_g}{\sqrt{1+\delta^2}+[(U_c-U_s)/2]\chi_g\pm\sqrt{U_a^2\chi_g^2+\delta^2}} \; , \qquad (20)$$

and the corresponding "normal" components are

$$M_- = \frac{1}{N}\left[M_3 - \frac{1}{\delta}(U_a\chi_g - \sqrt{U_a^2\chi_g^2+\delta^2})M_4\right] \; ,$$

$$\qquad (21)$$

$$M_+ = \frac{1}{N}\left[\frac{1}{\delta}(U_a\chi_g - \sqrt{U_a^2\chi_g^2+\delta^2})M_3 + M_4\right] \; ,$$

with

$$N \equiv (\sqrt{2}/\delta)\sqrt{\delta^2+U_a^2\chi_g^2-U_a\chi_g\sqrt{U_a^2\chi_g^2+\delta^2}}$$

and $U_a \equiv (U_c+U_s)/2$.

C. Landau free-energy expansion

The susceptibility (18) determines completely the quadratic part of the free energy

$$F^{(2)} = \frac{ab}{(2\pi)^2}\int d^2q\,[M_x^\dagger(\mathbf{q})M_y^\dagger(\mathbf{q})M_z^\dagger(\mathbf{q})C^\dagger(\mathbf{q})]\chi^{-1}(\mathbf{q},\omega=0) \begin{bmatrix} M_x(\mathbf{q}) \\ M_y(\mathbf{q}) \\ M_z(\mathbf{q}) \\ C(\mathbf{q}) \end{bmatrix} \qquad (22)$$

with the inverse susceptibility matrix given by

$$[\chi^{-1}]_{ij} = \begin{vmatrix} \dfrac{1-U_s\chi_0}{\chi_0} & 0 & 0 & 0 \\[3mm] 0 & \dfrac{1-U_s\chi_0}{\chi_0} & 0 & 0 \\[3mm] 0 & 0 & \dfrac{\sqrt{1+\delta^2}-U_s\chi_g}{\chi_g} & -\dfrac{\delta}{\chi_g} \\[3mm] 0 & 0 & -\dfrac{\delta}{\chi_g} & \dfrac{\sqrt{1+\delta^2}+U_c\chi_g}{\chi_g} \end{vmatrix} \; . \qquad (23)$$

In order to extend the Landau expansion to the ordered state below critical temperature we have to include the fourth-order terms in M_i. Expecting that the main effects of both Pauli and orbital couplings are present in the quadratic term (22), we use the result for the fourth-order SDW term obtained by Hasegawa and Fukuyama:[20]

$$F^{(4)} = \frac{B}{2} \int dx [2(|\mathbf{M}|^2)^2 - \mathbf{M}^{*2}\mathbf{M}^2] , \qquad (24)$$

where $B \approx n_F U_s^4 \zeta(3)/4(\pi T)^2$, with n_F being the density of states at the Fermi level. This extension is appropriate for the repulsive interactions $U_s, U_c > 0$ which favor the SDW ordering, and not the CDW one. Then the coefficients in front of $|M_i|^2$, $i = 1, 2, 3$ in Eq. (22) are all close to zero and are therefore critical, while the coefficient χ_{44}^{-1} remains large at all temperatures of interest. Thus, M_4 is linearly driven by M_3 through the bilinear coupling $[\chi^{-1}]_{34} = [\chi^{-1}]_{43}$ so that

$$M_4(\mathbf{q}) = \frac{\delta(\mathbf{q})}{\sqrt{U_a^2 \chi_g^2(\mathbf{q}) + \delta^2(\mathbf{q})} + U_a \chi_g(\mathbf{q})} M_3(\mathbf{q})$$

$$\approx \frac{\delta(\mathbf{q})}{2 U_a \chi_g(\mathbf{q})} M_3(\mathbf{q}) . \qquad (25)$$

The second equality in Eq. (25) gives the SDW$_\parallel$-CDW hybridization ratio M_3/M_4 in the limit of small δ. [We get the same expression in the fluctuation regime $(T > T_c)$, expanding the constraint $M_+ = 0$ in terms of small δ.] Note that there is no hybridization at $\mathbf{q} = 0$ since $\delta(\mathbf{q} = 0) = 0$. Using relation (25) we eliminate M_4 from $F^{(2)}$, so that it reduces to

$$F^{(2)} = \frac{ab}{(2\pi)^2} \int d\mathbf{q} [D_\perp(\mathbf{q})|M_\perp(\mathbf{q})|^2 + D_\parallel(\mathbf{q})|M_\parallel(\mathbf{q})|^2] \qquad (26)$$

with $M_\parallel \equiv M_3$ and

$$D_\perp(\mathbf{q}) = \chi_0^{-1}(\mathbf{q}) - U_s , \qquad (27)$$

$$D_\parallel(\mathbf{q}) = \frac{(\chi_g^{-1} + U_c)(\chi_g^{-1} - U_s)}{\chi_g^{-1}\sqrt{1 + \delta^2} + U_c} . \qquad (28)$$

The thermodynamics of the ordered SDW in the external magnetic field and in the temperature range below the critical temperature is thus described by the Landau free-energy functional $F\{M\} = F^{(2)} + F^{(4)}$, with $F^{(2)}$ and $F^{(4)}$ given by (26) and (24), respectively.

III. MEAN-FIELD CRITICAL TEMPERATURE

In the mean-field approximation one retains only the Fourier components $M_\perp(\kappa_\perp)$ and $M_\parallel(\kappa_\parallel)$ with wave vectors $\kappa_\perp$ and $\kappa_\parallel$ for which the respective coefficients $D_\perp$ and $D_\parallel$ in Eq. (26) are minimal. It is easy to show that one then has in the ordered state either $M_\perp \neq 0$ $M_\parallel = 0$ or $M_\perp = 0$ $M_\parallel \neq 0$, for $D_\perp(\kappa_\perp) < D_\parallel(\kappa_\parallel)$ and $D_\perp(\kappa_\perp) > D_\parallel(\kappa_\parallel)$, respectively. Thus, SDW can be oriented either perpendicularly or parallelly to the magnetic field. We show below that although the SDW in a purely isotropic sys-

tem orders perpendicularly $(M_\perp \neq 0, M_\parallel = 0)$ in the whole temperature range below the critical temperature, the finite internal spin anisotropy may cause a more complex behavior.

The critical temperature $T_c^\perp$ for the SDW component perpendicular to the magnetic field (SDW$_\perp$) follows from the equation

$$\chi_{11,22}^{-1}(\omega = 0) = 0 . \qquad (29)$$

Analogously, the critical temperature $T_c^\parallel$ for the hybridized parallel order (SDW$_\parallel$) is determined by

$$\chi_-^{-1}(\omega = 0) = 0 . \qquad (30)$$

It is easy to see that for weak magnetic fields $(h \equiv \mu_B H/2\pi T \ll 1)$ $T_c^\parallel < T_c^\perp$. Namely, expanding the denominator of χ_- in terms of h up to the second order, one gets

$$T_c^\parallel \approx T_c^\perp \exp[-7\zeta(3)h^2] . \qquad (31)$$

In this range the wave vectors of ordering of SDW$_\perp$ and SDW$_\parallel$ coincide, i.e., $\kappa_\perp = \kappa_\parallel = 0$. Relation (31) is independent on t_b. It is also almost independent of ε_0 as long as $\varepsilon_0 \le 0.5\Delta_0$, where Δ_0 is the gap at $T = 0$ for the perfect nesting. The numerical extension of the relation (31) to larger values of magnetic field is shown in Fig. 1. $T_c^\parallel/T_c^\perp$ continues to decrease as H increases. However, as is shown in the inset of Fig. 1, above a given critical value of magnetic field h_c the longitudinal component of $\kappa_\parallel$ shifts from zero to finite values $\pm \kappa_{\parallel a}(h)$, which are proportional to $(h - h_c)^{1/2}$ for $h - h_c \ll 1$, and have the asymptotic behavior $|\kappa_{\parallel a}(h)| \approx 4\pi Th/v_F$ for large values of $h - h_c$. The value of h_c is also independent on t_b and equal to 0.34 for $\varepsilon = 0$. It weakly decreases as ε_0 increases. Furthermore, for $\eta \le 1.55$ the transverse component of $\kappa_\parallel$ also shifts from zero. Small values of η may be realized by directing $\mathbf{H}$ closely to $\mathbf{j}$. For example, $\eta \le 1.55$ corresponds to $\theta \ge 52°$ at $h \approx h_c$ in Bechgaard salts. The shift of $\kappa_{\parallel 2}$, is, however, not followed by the
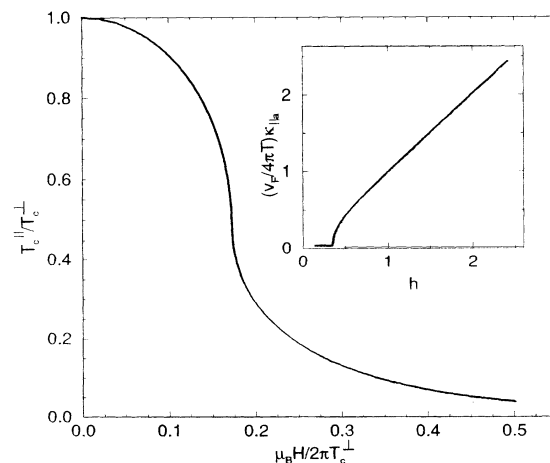

FIG. 1. The dependence of the ratio $T_c^\parallel/T_c^\perp$ on magnetic field for $\varepsilon_0 = 0$ and $\eta > 1.55$. The inset represents the dependence of $(v_F/4\pi T)\kappa_{\parallel a}$ on the h for the same choice of ε_0 and η.

substantial change of the $T_c^{\parallel}(H)$ dependence shown in Fig. 1. From now on we shall limit the discussion to the range $\eta > 1.55$.

The phase $SDW_\perp$ remains stable also at $T < T_c^{\parallel}$, i.e., it cannot be replaced by the phase $SDW_\parallel$ at some lower temperature. Namely, it is easy to see from Eqs. (27) and (28) that the absolute minimum of $D_\perp$ is lower than the absolute minimum of $D_\parallel$ for any fixed values of H and T. Thus, we conclude that the Zeeman splitting has no effect on the mean-field critical temperature and cannot induce any phase transition below this temperature.

In the presence of internal spin anisotropy the SDW order parameter $\langle \mathbf{M} \rangle$ is at $\mathbf{H}=0$ oriented along an easy magnetic axis. If the external magnetic field is directed in this direction, the SDW will change the orientation by $\pi/2$ at some finite value of H. In order to derive the expression for this spin-flop field within the present mean-field approach we adopt a simplified starting point which is not far from the real situation[21] in the Bechgard salts. Assuming that the easy spin direction is along the b axis and other two, intermediate and heavy, directions coincide with the a and c axes, respectively,[22] we write the interaction part of the Hamiltonian in the form

$$\mathbf{H}_{int} = -\frac{1}{2} \sum_{\mathbf{R}_\perp} \int dx [U_b (M_b^\dagger M_b + M_b M_b^\dagger)$$

$$+ U_a (M_a^\dagger M_a + M_a M_a^\dagger)] . \qquad (32)$$

Here U_b and U_a are coupling constants which presumably include the effective contributions from the dipole-dipole and spin-orbit interactions. M_b and M_a are now SDW components in the b and a directions, respectively. The third (c) component of $\mathbf{M}$ is irrelevant for further discussion. We also assume that $h < h_c$, so that $\kappa_\perp = \kappa_\parallel$. Then there is no SDW-CDW hybridization, i.e., $\delta = 0$ in Eq. (25) so that the CDW component M_4 can be omitted from calculations.

The calculation equivalent to that in Sec. II leads to the quadratic term in the Landau expansion

$$F^{(2)} = n_F \left[U_b^2 \ln \frac{T}{T_c^b} |M_b|^2 + U_a^2 \ln \frac{T}{T_c^a} |M_a|^2 \right] , \qquad (33)$$

where T_c^a and T_c^b are mean-field critical temperatures for the SDW order along the intermediate and easy axes, respectively,

$$T_c^a \approx 2\eta E_F / \pi \exp[-1/n_F U_a] ,$$

$$T_c^b \approx T_c^a \exp \left[\frac{1}{n_F} \left[\frac{1}{U_a} - \frac{1}{U_b} \right] - 7\zeta(3)h^2 \right] . \qquad (34)$$

With result (30) instead of (31), it comes out that the SDW order parallel to the easy axis (and to the external magnetic field) remains stable until a spin-flop field given by

$$h_{sf} = \left[\frac{1}{7\zeta(3)n_F} \left[\frac{1}{U_a} - \frac{1}{U_b} \right] \right]^{1/2} . \qquad (35)$$

For $h < h_{sf}$ the critical temperature depends on h as specified by Eq. (31). For $h > h_{sf}$ the SDW is oriented perpendicularly to $\mathbf{H}$, and the critical temperature ceases

to be field dependent.

The experimental values of spin-flop field $(H_{sf} \approx 0.5$ T$)$ (Ref. 22) and critical temperature $(T_c \approx 10$ T$)$ for Bechgaard salts situate the spin-flop transition in the range $h \approx h_{sf} \ll h_c$, which justifies the initial assumptions in Eq. (32). The present mean-field evaluation of spin-flop transition is complement to that via the bosonization and renormalization procedure performed by Giamarchi and Schulz.[21]

IV. FLUCTUATIONS

In the discussion of the effects of the magnetic field on the Gaussian SDW fluctuations we shall consider separately the temperature ranges closely above and below T_c. It should be noted that due to the Landau damping all SDW collective modes are diffusive in both regimes, so that one cannot speak about dispersive branches. We therefore consider the components χ_{11}, χ_{22}, and χ_- of the diagonal susceptibility matrix, which are propagators for fluctuations of M_1, M_2, and M_-, respectively. We omit the component M_+ which is not critical, and take $U_s = U_c = U$ for simplicity.

A. $T \geq T_c$

In the absence of external magnetic field and internal spin anisotropy the component χ_- reduces to χ_{33} and becomes degenerate to χ_{11} and χ_{22}. Then all six modes corresponding to fluctuations of real and imaginary parts of M_1, M_2, and M_3 are degenerated. The external magnetic field splits this degeneracy through the Pauli coupling into four modes corresponding to perpendicular (i.e., χ_{11} and χ_{22}) fluctuations and two modes corresponding to parallel (i.e., χ_-) fluctuations.

The expansion of the propagators around their maxima up to the quadratic power in the wave vector and the linear power in the Matsubara frequency[23] gives

$$\chi_{11}(\mathbf{q}, \omega_n) = \chi_{22}(\mathbf{q}, \omega_n)$$

$$\approx n_F^{-1} U^{-2} \left[a_\perp + \xi_{1a}^2 q_1^2 + \xi_{1b}^2 q_2^2 - \frac{\pi}{8T} \alpha_\perp \omega_n \right]^{-1}$$

$$\qquad (36)$$

and

$$\chi_-(\mathbf{q}, \omega_n)$$

$$\approx n_F^{-1} U^{-2} \left[a_\parallel + \xi_{\parallel a}^2 q_1^2 + \xi_{\parallel b}^2 q_2^2 - \frac{\pi}{8T} \alpha_\parallel \omega_n \right]^{-1} . \qquad (37)$$

Coefficients $a_{\perp(\parallel)}$ vanish at temperatures $T_c^{\perp(\parallel)}$ considered in Sec. III, and vary linearly with temperature closely to them. Coefficients $\xi_{1a,b}$ and $\xi_{\parallel a,b}$ are the respective correlation lengths in the longitudinal (a) and transverse (b) directions, while $\alpha_\perp$ and $\alpha_\parallel$ are corresponding damping parameters. The field (i.e., h) dependence of these coefficients is illustrated in Figs. 2–4 for $n_F U = 0.2$, $t_b = 300$ K, and two values of the parameter of imperfect nesting, i.e., for the perfect $(\varepsilon_0 = 0)$ and bad $(\varepsilon_0 = 0.95)$ nesting, at the temperature $T = T_c^\perp (\varepsilon_0 = 0) \approx 10.5$ K. For
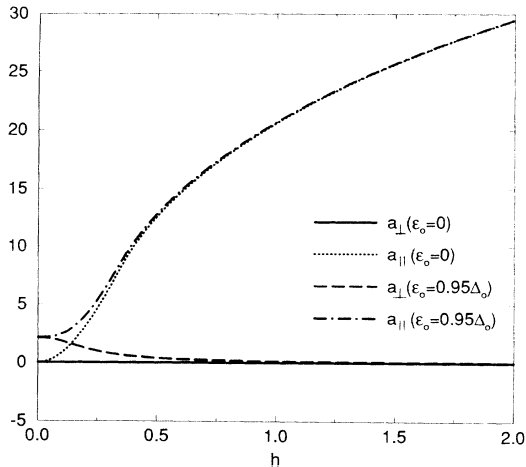
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FIG. 2. The coefficients $a_\perp$ and $a_\parallel$ as functions of h at $T = T_c^\perp$ ($\varepsilon_0 = 0$), for perfect and bad nesting.

both values of ε_0 the equilibrium SDW wave vector at $h = 0$ is equal to zero, i.e., one does not have the stabilization of the modulation different from that of perfect nesting.[24,25]

As is clear from Fig. 2, the difference $a_\parallel - a_\perp$ increases for both perfect and bad nesting as the magnetic field increases. At small values of h this difference is parabolic in h, as follows from the result (31) after taking into account that $a_{\perp,\parallel} \approx \ln T / T_c^{\perp,\parallel}$. In the case $\varepsilon_0 \neq 0$, the orbital effects cause the decrease of $a_\perp$ as h increases, i.e., one recovers the already known[26] increase of the critical temperature $T_c^\perp$. Due to the increase of $a_\parallel(h)$, the spectral weight of M_- fluctuative modes shifts to higher frequencies, i.e., these fluctuations are less and less critical as H increases. Note that the shift of the wave number $\kappa_{\parallel a}$ from 0 to finite values at $h \geq h_c$ does not cause any anomaly in the $a_\parallel(h)$ dependence.

The longitudinal and transverse correlation lengths are shown in Figs. 3(a) and 3(b), respectively. As is already known,[14] the orbital coupling at $\varepsilon_0 = 0$ does not affect the longitudinal correlation length ξ_{1a}, but causes a decrease of the transverse correlation length ξ_{1b}. For $\varepsilon_0 \neq 0$, ξ_{1a} increases with h and finally saturates at its value for the perfect nesting. Similarly, the dependence of ξ_{1b} on h turns, after an initial increase, to the law which ap-
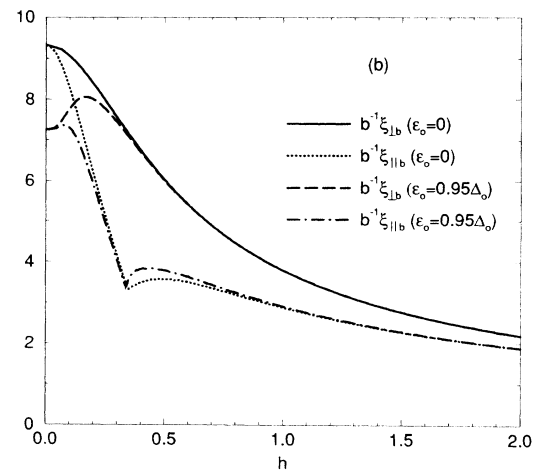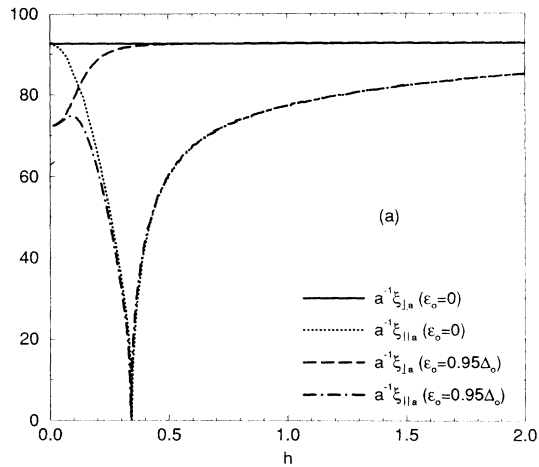
FIG. 3. (a) The dependence of longitudinal and (b) transverse correlation lengths (measured in terms of respective lattice constants) on h at $T = T_c^\perp$ ($\varepsilon_0 = 0$) for perfect and bad nesting. [$t_b = 300$ K.]

proaches the curve $\xi_{1b}(h)$ for the perfect nesting. The magnetic field thus effectively improves the nesting through the orbital coupling, in accordance with the general propositions.[9]

The Pauli coupling affects only $\xi_{\parallel a}$ and $\xi_{\parallel b}$ in Fig. 3, causing an additional decrease of both correlation lengths as h increases. This decrease starts quadratically in h. The corresponding expansions for $\varepsilon_0 = 0$ are

$$\xi_{\parallel a}^2 \approx \frac{v_F^2 7 \zeta(3)}{(4\pi T)^2} \left[1 - 2 \left[\frac{93 \zeta(5)}{7 \zeta(3)} - 14 n_F U \zeta(3) \right] h^2 \right] , \tag{38}$$

$$\xi_{\parallel b}^2 \approx \left[\frac{t_b}{4\pi T} \right]^2 b^2 14 \zeta(3) \left\{ 1 - \left[\frac{31 \zeta(5)}{7 \zeta(3)} \left[6 + \frac{\eta^2}{4} \right] - 14 n_F U \zeta(3) \right] h^2 \right\} . \tag{39}$$

The orbital coupling enters through the parameter η defined in the Introduction. Both expansions also contain the terms proportional to $n_F U$, which are usually omitted in the weak-coupling limit. Equation (39) reduces to the previous result[14] for the orbital contribution to ξ_{1b} after neglecting the Pauli contribution (by put-

ting, e.g., $\mu_b \to 0$). The decrease of $\xi_{\parallel a}$ and $\xi_{\parallel b}$ as h increases is connected with the formation of the new shifted maxima of χ_- for $h > h_c$. At $h = h_c$, $\xi_{\parallel a}$ attains zero, while $\xi_{\parallel b}$ attains minimum. The values of $\xi_{\parallel a}$ and $\xi_{\parallel b}$ at $h > h_c$ correspond to the expansion around the new maxima of χ_-.
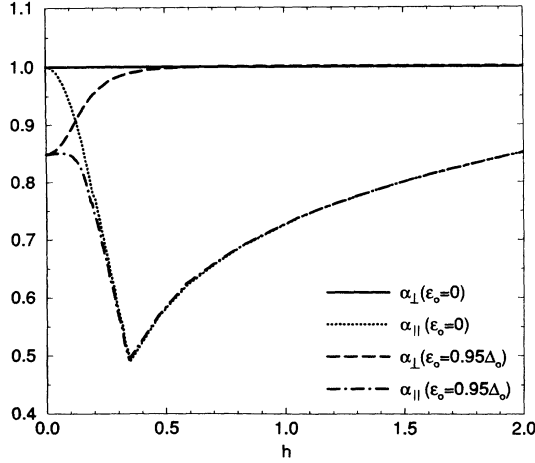
FIG. 4. The attenuation coefficients $\alpha_\perp$ and $\alpha_\parallel$ as functions of h at $T = T_c^\perp (\varepsilon_0 = 0)$ for perfect and bad nesting.

The field dependence of the damping constants $\alpha_\perp$ and $\alpha_\parallel$ (Fig. 4) is similar to that of the corresponding longitudinal correlation lengths. At $h = h_c$, $\alpha_\parallel$ does not reach zero and has a jump in the slope instead of the analytic minimum. The expansion of $\alpha^\parallel$ in terms of h for the case of perfect nesting is given by

$$\alpha_\parallel \approx 1 - [\pi^2 - 14 n_F U \zeta(3)] h^2 . \tag{40}$$

B. $T < T_c$

The order parameter for the SDW is six dimensional. The setin of order in an isotropic system fixes three variables, while the remaining three variables are continuously degenerate. The latter correspond to free translations of SDW and to the freedom in the orientation of $\langle \mathbf{M} \rangle$. With the anisotropy due to the finite magnetic field the

degeneracy is reduced to two dimensions since the rotations of $\langle \mathbf{M} \rangle$ are limited to the plane perpendicular to $\mathbf{H}$. Finally, the internal spin anisotropy generally imposes the preferable direction of $\langle \mathbf{M} \rangle$ so that there remains only the translational degeneracy.

Together with the above breaking of symmetry, the SDW order also induces further splitting among six fluctuative modes. The number of gapless (Goldstone) modes coincide with the dimension of degeneracy. In order to specify those as well as remaining modes with finite gaps for $\mathbf{H} = 0$ and $\mathbf{H} \neq 0$ in an isotropic system, we choose a convenient set of six coordinates and then derive the free-energy functional at $T < T_c$ with all harmonic fluctuative contributions taken into account. We represent three complex components of $\mathbf{M}$ by

$$M_1 = m_1 e^{i\phi} ,$$
$$M_2 = (m_2 + i\tau_2) e^{i\phi} , \tag{41}$$
$$M_3 = (m_3 + i\tau_3) e^{i\phi} ,$$

where all variables on the right-hand side are real by definition. It is easy to see by minimizing the Landau expansion (24) and (26) that the ordered SDW state for the isotropic case is defined by

$$m_1^2 + m_2^2 + m_3^2 \equiv M_0^2 = -\frac{a}{B} , \quad \tau_{20} = \tau_{30} = 0 , \tag{42}$$

with $a \equiv n_F U^2 \ln T / T_c$. Thus, all three components have a common phase, i.e., the SDW is a linearly polarized $2k_F$ wave.[27,28]

Using the polar coordinates instead of "Cartesian" ones ($m_1 = M \sin\theta \cos\varphi$, $m_2 = M \sin\theta \sin\varphi$, $m_3 = M \cos\theta$) and making the gradient expansion and the harmonic expansion in terms of nondegenerate variables τ_2, τ_3 and $M \equiv M_0 + \delta$, one gets the free-energy functional for fluctuations

$$\delta F = \int d^2 x \left[(-2a)[\delta^2 + (1 + \cos^2\theta \sin^2\theta)\tau_2^2 + \sin^2\theta \tau_3^2 - \sin2\theta \sin\varphi \tau_1 \tau_2] \right.$$

$$+ \sum_{i=a,b} \xi_i^2 \{ (\partial_i \delta)^2 + (\partial_i \tau_2)^2 + (\partial_i \tau_3)^2 + M_0^2 [(\partial_i \theta)^2 + \sin^2\theta (\partial_i \varphi)^2 + (\partial_i \phi)^2]$$

$$\left. + 2 M_0 (\sin\theta \sin\varphi \, \partial_i \tau_2 + \cos\theta \, \partial_i \tau_3) \partial_i \phi \} \right] . \tag{43}$$

This functional is, except for the fluctuations of total amplitude (δ), neither linear nor diagonal. However, a complete separation and linearization is achieved after making the usual assumption that there are no strong angular variations, i.e., that the direction of $\mathbf{M}$ does not change at scales smaller and comparable to that of long-wavelength limit. Let us put, e.g., $\theta_0 = \pi/2$, $\varphi_0 = 0$ (i.e., $m_{10} \neq 0$, $m_{20} = m_{30} = 0$). One immediately recognizes three gapless modes with the same coefficient of elasticity for the phason (ϕ) and two spin waves ($\varphi, \theta - \pi/2$), and three modes with the same gap corresponding to the amplitudon (δ) and the fluctuations of relative phases of complex components out of M_1 (τ_2 and τ_3).

In the presence of the finite magnetic field the ordered state is defined by $\tau_{20} = \tau_{30} = m_{30} = 0$ and $m_1^2 + m_2^2 \equiv M_0^2 = -a_\perp / B$, providing that both $D_\perp$ and $D_\parallel$ in Eq. (26) have minima at the common wave vector $\kappa_\perp = \kappa_\parallel = 0$. Here, as before, we put the component M_3 in the direction of $\mathbf{H}$. After choosing cylindrical coordinates ($m_1 = M \cos\varphi$, $m_2 = M \sin\varphi$, m_3), making the harmonic expansion in terms of $M = M_0 + \delta$, m_3, τ_2, and τ_3, and fixing the direction $\langle \mathbf{M} \rangle$ by choosing $\varphi_0 = 0$, the free-energy functional for fluctuations reduces to

$$\delta F = \int d^2x \left[(-2a_\perp)(\delta^2 + \tau_2^2) + (-3a_\perp + a_\parallel)\tau_3^2 + (a_\parallel - a_\perp)m_3^2 \right.$$

$$\left. + \sum_{ab} \xi_{1i}^2 \{ (\partial_i \delta)^2 + (\partial_i \tau_2)^2 + M_0^2[(\partial_i \varphi)^2 + (\partial_i \phi)^2] \} + \xi_{\parallel i}^2[(\partial_i m_3)^2 + (\partial_i \tau_3)^2] \right] . \tag{44}$$

As is seen from this expression, the Pauli coupling does not affect two Goldstone modes, i.e., the phason (ϕ) and the fluctuations of the SDW orientation in the plane perpendicular to H (φ). The same is valid for two massive modes limited to this plane, i.e., for the fluctuations of the amplitude (δ) and of the relative phase associated to τ_2. The remaining two massive modes are sensitive to the Pauli coupling. It has the most important effect on the m_3 mode, i.e., on fluctuations of the SDW orientation out of (M_1, M_2) plane. This mode acquires a small gap $(a_\parallel - a_\perp)^{1/2}$ which vanishes as H→0, as is shown in Fig. 2. The mode τ_3 representing fluctuations of relative phase of complex component M_3 with respect to ϕ, depends on the Pauli coupling only weakly, through a small increase of the gap from $(-2a_\perp)^{1/2}$ to $[-2a_\perp + (a_\parallel - a_\perp)]^{1/2}$. At higher magnetic fields when $\kappa_\parallel$ splits from $\kappa_\perp = 0$ these two modes are however, less distinctive. In particular, when $\kappa_\parallel$ is far enough from $\kappa_\perp$ so that the integrals of terms in $F^{(4)}$ [Eq. (24)] comprising oscillatory factors $\exp[\pm 2i(\kappa_\perp - \kappa_\parallel)\cdot\mathbf{x}]$ are negligible, they become degenerate, with a common gap $(-2a_\perp + a_\parallel)^{1/2}$.

Finally, the Pauli coupling also renormalizes the dispersions of both m_3 and τ_3 modes through the additional H dependence of $\xi_{\parallel a}$ and $\xi_{\parallel b}$ shown in Figs. 3(a) and 3(b). We remind the reader that all six modes are affected by the orbital coupling which enters in both ($\perp$) and ($\parallel$) coefficients in the expansion (44).

V. CONCLUSIONS

The most important effect of the Pauli coupling to CDW's is an additional breaking of symmetry of its six-dimensional order parameter. In systems with internal spin isotropy this does not cause any change in the critical temperature like in the case of CDW's,[15,16] but has as the main effect the splitting of two from remaining four fluctuative modes. The hardening of these two modes above T_c and opening of the small gap in one of three Goldstone modes below T_c are expected to have direct consequences on the measurable properties like magnetoresistance, specific heat, etc. The quantitative analysis of these effects will be presented elsewhere.

The effects of Pauli coupling on critical fluctuations are, in contrast to those of orbital coupling, independent of the direction of H. It is therefore possible to separate two types of effects by comparing the field dependence of experimental data for various directions of H. For exam-

ple, by directing H in the transverse b direction in the $(TMTSF)_2X$ salts one excludes to a great extent the orbital mechanism, so that the remainder field dependence is to be associated mainly to the Pauli mechanism, particularly at magnetic fields stronger than the spin-flop field.

The driving of the CDW by the component of the SDW parallel to H is another phenomenon linked to the Pauli mechanism. Since $SDW_\parallel$ is not ordered (except in the case of anisotropic system with H directed along the easy axis and weaker than the spin-flop field), this driving induces only CDW fluctuations at wave vectors at which the off-diagonal coupling (25) is finite. The range of such wave vectors widens as H increases. In particular, for $h > h_c$ the $CDW_\parallel$-CDW hybridization becomes stronger due to the shift of $\kappa_\parallel$ from zero (inset in Fig. 1). The rough estimation of the CDW fraction in this hybridization gives values of 10%–25 % as h runs from 0.4 to 1. These values are only weakly sensitive on imperfect nesting ε_0. With the finite contribution of CDW fluctuations one also induces fluctuations of underlying lattice. The Pauli coupling is therefore a possible mechanism for finite magnetoelastic effects mediated by $SDW_\parallel$ fluctuations.

Aside from the separable contributions, expressions (24) and (26) for the Landau expansion also contains terms which are of the mixed, Pauli, and orbital origin. The most interesting among these contain a new type of angular resonances, besides those coming from the sole orbital coupling.[13] They take place at directions of H defined by $\eta = 4/n$ (e.g., $\cos\theta = 4/\mu_B/v_F ebn$) where n is an integer, and have the strongest effect on the longitudinal correlation length of collective modes affected by Pauli coupling. At some of these angles $\xi_{\parallel a}$ shows picks, which are wider and wider as temperature increases.[29] The same type of resonances in the metallic state were found by Lebed[30] within Yakovenkos[31] approach to orbital angular resonances.

Finally, we stress again the reach content of the order-parameter space for SDW's, and the importance of splittings in this space induced by the Zeeman splitting. In this respect the topological defects in the SDW and their role in the nonlinear collective transport are questions which remain for future investigations.

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