

Spin-anisotropic electron-electron interactions in one-dimensional metals

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We construct a theoretical model for one-dimensional conductors taking into account the effects of spin-orbit coupling and electron-electron dipole interaction. New types of electron-electron interactions which do not conserve the z component of spin are generated. Use of bosonization and renormalization allows us to obtain correlation functions and the phase diagram. The main result is the appearance of a gap in the spin-excitation spectrum in the whole phase diagram, which leads to anisotropic spin-density-wave and superconducting phases. We discuss possible experimental implications of our results.

The quasi-one-dimensional organic conductors¹ with tetramethyltetraselenafulvalene (TMTSF) or tetramethyltetrathiafulvalene (TMTTF) chains, e.g., (TMTSF)₂X or (TMTTF)₂X, exhibit in many cases low-temperature phase transitions from a metallic into an antiferromagnetically ordered insulating state.²⁻⁴ Quite generally, this state shows easy-axis-type anisotropy, with the easy axis along the crystalline b direction (perpendicular to the conducting chains).⁴ There is also anisotropy in the plane perpendicular to the easy axis. Clearly the anisotropic antiferromagnetism must be due to spin-space anisotropic electron-electron interactions and thus cannot be explained by Coulomb and electron-phonon interactions alone.

The theoretical description of those substances is often based on the one-dimensional interacting electron-gas model.⁵⁻⁷ One frequently introduces anisotropic spin interactions, however, without specifying the origin of the anisotropy. We try here to give a physical meaning to such an anisotropy by taking spin-orbit coupling⁸ (SOC) and electronic dipole-dipole interactions (DD) into account. We first derive the anisotropic electron-electron coupling constants generated by such an interaction. In addition to the usually considered anisotropies we find spin-nonconserving couplings not previously considered. A renormalization-group analysis shows that these couplings lead to some qualitative changes in the standard picture of the one-dimensional electron gas. We finally give a brief discussion of possible experimental implications.

For definiteness, consider a chain of molecules whose levels are, without SOC, only Kramers degenerate. The chain axis is along z , and the conduction band is built out of p_z -like orbitals. To have an idea of the effect of SOC let us consider a model molecule with three nondegenerate orbitals \tilde{p}_j which have the same symmetry properties as p_x, p_y, p_z orbitals. Diagonalization of the molecular Hamiltonian gives, to first order in the SOC, the eigenfunctions

$$\begin{pmatrix} \phi \\ \psi \end{pmatrix} = |\Phi + \rangle = |\tilde{p}_z \uparrow \rangle + A |\tilde{p}_x \downarrow \rangle - iB |\tilde{p}_y \downarrow \rangle, \quad (1)$$

$$K |\Phi + \rangle = |\Phi - \rangle = -A |\tilde{p}_x \uparrow \rangle + iB |\tilde{p}_y \uparrow \rangle + |\tilde{p}_z \downarrow \rangle,$$

with

$$E_i = \langle \tilde{p}_i | H_0 | \tilde{p}_i \rangle,$$

$$A = (E_x - E_z)^{-1} \langle \tilde{p}_x \uparrow | \alpha \mathbf{L} \cdot \mathbf{S} | \tilde{p}_z \downarrow \rangle, \quad (2)$$

$$iB = (E_y - E_z)^{-1} \langle \tilde{p}_z \uparrow | \alpha \mathbf{L} \cdot \mathbf{S} | \tilde{p}_y \downarrow \rangle.$$

Here K is the time inversion operator, $|\uparrow \downarrow \rangle$ are the eigen-

functions of S_z , and for $\alpha \rightarrow 0$ one has $S_z |\pm \rangle = \pm |\pm \rangle$. The x axis will be chosen in the direction so that $|A/B|$ is maximum. As in TMTSF, coupling is expected to be stronger in the longitudinal direction, we will choose this as x axis; this ensures that ψ is nearly real. Assuming inversion symmetry of the crystals, the tight-binding Bloch functions are

$$|k + \rangle = \begin{pmatrix} \phi_k \\ \psi_k \end{pmatrix} = N^{-1/2} \sum_l e^{ikl} \begin{pmatrix} \phi(z-l) \\ \psi(z-l) \end{pmatrix}, \quad |k - \rangle = \begin{pmatrix} -\psi_{-k}^* \\ \phi_{-k}^* \end{pmatrix}. \quad (3)$$

States $|\pm k, \pm \rangle$ are degenerate. In the absence of inversion symmetry this degeneracy is lifted, but this case will not be considered here.

We assume that the electrons interact through a spin-independent potential. Then $|\pm \rangle$ are no longer conserved because they are not spin eigenvectors. As usual we are only interested in processes near the Fermi surface. We consider effective short-range interaction which we parametrize using the standard g -ology notation.⁵ In addition to the standard interactions we find the *new diagrams* shown in Fig. 1. These terms do not conserve the z component of the effective spin $|\pm \rangle$. The g_4 process leads only to a renormalized Fermi velocity and will not be considered here. Other diagrams generated by SOC are zero because of time-reversal symmetry.

Explicit expressions for the anisotropic interactions are

$$(g_{1\parallel} - g_{1\perp})_{\text{SO}} = -V(q=0) \int dx \|\psi_{k_f} \phi_{-k_f} - \psi_{-k_f} \phi_{k_f}\|^2,$$

$$(g_f)_{\text{SO}} = -V(q=0) \text{Re} \int dx (\psi_{k_f} \phi_{-k_f} - \psi_{-k_f} \phi_{k_f})^2,$$

$$(g_c)_{\text{SO}} = -V(q=0) \text{Im} \int dx (\psi_{k_f} \phi_{-k_f} - \psi_{-k_f} \phi_{k_f}) \phi_{k_f} \phi_{-k_f}. \quad (4)$$

At this order we have still $g_{2\perp} = g_{2\parallel}$, and for a repulsive interaction and $|A| \gg |B|$,

$$(g_{1\parallel} - g_{1\perp})_{\text{SO}} \approx -(g_f)_{\text{SO}} < 0. \quad (5)$$

Notice that $g_{1\parallel} - g_{1\perp}$ and g_f depend on the SOC squared, so at the same order of magnitude we have also to consider the magnetic dipole-dipole (DD) interaction between electrons. We limit the DD interaction to a single chain, thus neglecting its long-range nature. The long-range character is important in the case of ferromagnetism. However, in (quasi-) one-dimensional systems one is mainly concerned with antiferromagnetic instabilities. In this case the long-

range effect is canceled by the alternation of magnetic moments. The main effect is to introduce a short-range, but spin-anisotropic interaction,⁹ which on a single chain is

$$H_{DD} = (\mu_1 \mu_2 / r^3) - 3[(\mu_1 \mathbf{r})(\mu_2 \mathbf{r}) / r^5] . \quad (6)$$

The main contribution of H_{DD} is for the g_2 and g_f processes (g_1 is expected to be much smaller because of the $2k_f$ transferred momentum). With our choice of axes we have

$$\begin{aligned} (g_{2\parallel} - g_{2\perp})_{DD} &= \langle k_f +, -k_f + | H_{DD} | k_f +, -k_f + \rangle \\ &\quad - \langle +, - | | +, - \rangle < 0 , \\ g_f &= \langle k_f -, -k_f - | H_{DD} | k_f +, -k_f + \rangle < 0 . \end{aligned} \quad (7)$$

The new interaction Hamiltonian is

$$H_{\text{int}} = L^{-1} \sum_{\sigma} g_f \int_0^L dx \psi_{+, \sigma}^{\dagger}(x) \psi_{-, \sigma}^{\dagger}(x) \psi_{-, -\sigma}(x) \psi_{+, -\sigma}(x) + L^{-1} \sum_{\sigma_1, \sigma_2} g_c \int_0^L dx \psi_{+, \sigma_1}^{\dagger}(x) \psi_{-, \sigma_2}^{\dagger}(x) \psi_{-, -\sigma_2}(x) \psi_{+, \sigma_1}(x) + (1 \leftrightarrow 2) . \quad (8)$$

\pm denotes right- and left-going fermions and $\sigma = \pm$ indicates spin up and down. We use the boson representation^{10,11} of fermion operators, introduce charge (ρ) and spin (σ) density operators in the standard way,⁵ and define the phase fields:

$$\phi_{\nu}, \theta_{\nu} = \mp (i\pi/L) \sum_{p \neq 0} (1/p) e^{i p / 2 - i p x} [\nu_{+}(p) \pm \nu_{-}(p)] , \quad \nu = \rho \text{ or } \sigma , \quad (9)$$

and in $A, B = \dots$ the upper sign refers to A . The part of the Hamiltonian bilinear in the boson operators is diagonalized by the usual canonical transformation with parameter

$$e^{2\xi\nu} = \left(\frac{2\pi\nu + g_{\parallel}}{2\pi\nu - g_{\parallel}} \right)^{1/2} , \quad g_{\parallel}, g_{\parallel} = g_{1\parallel} - g_{2\parallel} \mp g_{2\perp}, \quad K_{\phi} = K_{\theta}^{-1} = e^{2\xi\sigma}, \quad K_{\rho} = e^{2\xi\rho} , \quad (10)$$

and by use of $\rho_{\pm}(x) = -(2\pi)^{-1} \nabla(\phi \mp \theta)$ the Hamiltonian reads

$$\begin{aligned} H &= H_{\rho} + H_{\sigma} + \frac{2g_{1\perp}}{(2\pi\alpha)^2} \int dx \cos\sqrt{8K_{\phi}} \phi_{\sigma} - \frac{2g_f}{(2\pi\alpha)^2} \int dx \cos\sqrt{8K_{\theta}} \theta_{\sigma} \\ &\quad + \frac{2g_c}{(2\pi\alpha)^2} \sum_{\pm} r \int dx \frac{-1}{2\pi} \nabla(e^{\xi\rho} \phi_{\rho} + r e^{-\xi\rho} \theta_{\rho}) \cos\sqrt{2}(K_{\phi}^{1/2} \phi_{\sigma} - r K_{\theta}^{1/2} \theta_{\sigma}) . \end{aligned} \quad (11)$$

H_{ρ} and H_{σ} are standard free-boson Hamiltonians.⁵ H is invariant under the duality transformation $\phi_{\sigma} \leftrightarrow \theta_{\sigma}$, $g_{1\perp} \leftrightarrow g_f$, $K_{\phi} \leftrightarrow 1/K_{\theta}$.

From a perturbation calculation of correlation functions we find the renormalization-group equations for a change of the length scale $\alpha \rightarrow e^l \alpha$. Introducing the reduced variables $g \leftarrow g/\pi\nu$ and expanding for small g 's we have

$$\begin{aligned} dg_{\parallel}/dl &= g_{\parallel}^2 - g_{\perp}^2 - (g_c^2 g_{\parallel}^2 / 2) , \\ dg_{1\perp}/dl &= -g_{1\perp} g_{\parallel} , \\ dg_f/dl &= g_f g_{\parallel} , \\ dg_c/dl &= -g_c g_{\parallel}^2 / 8 . \end{aligned} \quad (12)$$

If $g_c = 0$ the equations can be solved exactly. We omit algebraic details and discuss only the main features. There is a fixed line $|g_f| = |g_{\perp}|$ and $g_{\parallel} = 0$ and a critical plane $g_{\parallel}^2 - |g_{1\perp}| + |g_f| = 0$ separating the two behaviors $g_{\parallel}^2 \rightarrow \pm\infty$. As in the standard case ($g_f = 0$) $g_{\parallel}^2 \rightarrow -\infty$ corresponds to long-range order of ϕ_{σ} and an exponential decay of correlation functions like $\langle e^{i\phi(x)} e^{-i\phi(x)} \rangle$. Using the duality transformation $\phi_{\sigma} \leftrightarrow \theta_{\sigma}$ one finds that for $g_{\parallel}^2 \rightarrow +\infty$ there is long-range order in θ_{σ} and an exponential decay of correlation functions like $\langle e^{i\phi(x)} e^{-i\phi(x)} \rangle$. In both cases there is a gap Δ_{σ} in the spin-density excitation spectrum (or equivalently a finite correlation length). Near the critical

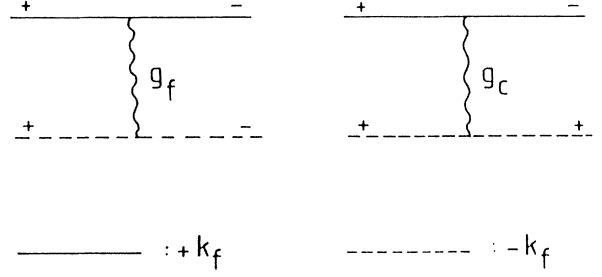


FIG. 1. New interactions introduced by spin-orbit or dipole-dipole coupling. Other diagrams are of order 1 or 2 in SOC and vanish at zero momentum transfer either by time-reversal symmetry or when antisymmetrized.

plane one has

$$\begin{aligned} \Delta_{\sigma} &\propto \delta^{\nu} , \\ \delta &= (4g_f g_{1\perp})^{-1} [g_{\parallel}^2 - (g_{1\perp} - g_f)^2] , \\ \nu &= (4|g_f g_{1\perp}|)^{-1/2} . \end{aligned} \quad (13)$$

We note that the exponent ν depends continuously on g_f and $g_{1\perp}$.

As can be seen from Eqs. (12) g_c is a marginal variable. If $g_c \neq 0$ the critical surface still exists but its shape and the critical exponents are changed. We still have two kinds of behavior: either long-range order of ϕ_{σ} or θ_{σ} .

The above analysis shows that, in contrast to previously studied cases, for nonzero g_f there is a spin-excitation gap in the whole parameter space (apart from the critical surface). To assess its influence on physical properties we investigate the different types of fluctuations⁵ [$2k_f$ charge-density wave (CDW), spin-density wave (SDW_i, $i = x, y, z$), or singlet (SS) and triplet Cooper pairings (TS)] typical of a one-dimensional metal. Specifically, we calculate the correlation functions:^{1,2}

$$R = -i\theta(t) \langle [O_i(r, t), O_i^{\dagger}(0, 0)] \rangle . \quad (14)$$

It is important to note the role played by the ϕ and θ fields in the different SDW _{α} and TS _{α} ($\alpha = x, y, z$) operators. We

have

$$\begin{aligned}
 O_{\text{CDW}} &= \frac{e^{2ik_f x}}{\pi\alpha} \exp[-i\sqrt{2}\phi_p(x)] \cos[\sqrt{2}\phi_\sigma(x)] , \\
 O_{\text{SDW}_x} &= \frac{e^{2ik_f x}}{\pi\alpha} \exp[-i\sqrt{2}\phi_p(x)] \cos[\sqrt{2}\theta_\sigma(x)] , \\
 O_{\text{SDW}_y} &= \frac{e^{2ik_f x}}{\pi\alpha} \exp[-i\sqrt{2}\phi_p(x)] \sin[\sqrt{2}\theta_\sigma(x)] , \\
 O_{\text{SDW}_z} &= \frac{e^{2ik_f x}}{\pi\alpha} \exp[-i\sqrt{2}\phi_p(x)] \sin[\sqrt{2}\phi_\sigma(x)] .
 \end{aligned} \quad (15)$$

O_{SS} and O_{TS} are obtained with $k_f = 0$ and replacing $\phi_p \rightarrow \theta_p$. The asymptotic behavior is $R_l(q, \omega) \sim \max(w, vq)^{-\alpha}$. The contribution of the charge part is easily obtained.⁵ For the spin part we notice that there is always a gap in the spin-excitation spectrum corresponding to a long-range order of ϕ or θ (respectively, for $g_{\parallel} \rightarrow -\infty$, $g_{\parallel} \rightarrow +\infty$). So we have

$$\begin{aligned}
 g_{\parallel} \rightarrow +\infty & \begin{cases} g_{1\perp} > 0 \phi_\sigma \rightarrow \pi/\sqrt{8} , \\ g_{1\perp} < 0 \phi_\sigma \rightarrow 0 , \end{cases} \\
 g_{\parallel} \rightarrow -\infty & \begin{cases} g_f < 0 \theta_\sigma \rightarrow \pi/\sqrt{8} , \\ g_f > 0 \theta_\sigma \rightarrow 0 . \end{cases}
 \end{aligned} \quad (16)$$

The exponents are only given by the charge part:

$$\alpha_{\text{CDW}}, \alpha_{\text{SDW}} = 2 - K_\rho, \quad \alpha_{\text{SS}}, \alpha_{\text{TS}} = 2 - K_\rho^{-1} . \quad (17)$$

But the correlation functions containing θ_σ (respectively, ϕ_σ) for $g_{1\perp} \rightarrow +\infty$ (respectively, $g_{1\perp} \rightarrow -\infty$) decay exponentially, so the divergence is removed. We are left with the phase diagrams in Fig. 2. On the critical surface we have $g_{\parallel} \rightarrow 0$, $|g_f| \rightarrow |g_{1\perp}|$, g_c not zero. All the exponents are zero, even if g_c is not zero as can be shown by a perturbation calculation.

The presence of the umklapp scattering in a half-filled band (g_3) leads to a gap in the density excitation spectrum⁵ and to long-range order in the ϕ_p field if $g_{\parallel} > 0$. Consequently, there are now (at $T=0$) *long-range ordered* SDW or CDW phases in the right part of the phase diagrams (Fig. 2).

If we interest ourselves in the weakly anisotropic limit (i.e., $g_{1\parallel} \approx g_{1\perp}$) there are two possibilities:

$$1. \quad g_{\parallel} < |g_{1\perp} - g_f| .$$

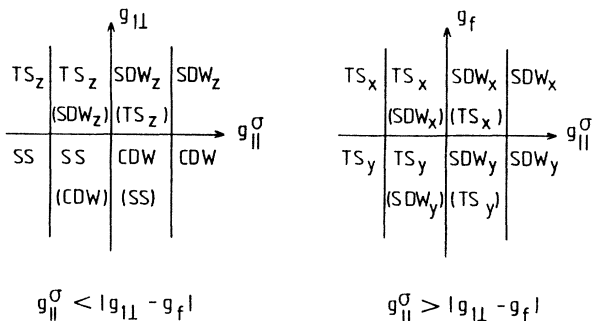


FIG. 2. Phase diagram. Parentheses indicate divergences weaker than the dominant one.

We are renormalized to $g_{\parallel} \rightarrow -\infty$ and find the usual CDW, SS phase diagram.⁵

$$2. \quad g_{\parallel} > |g_{1\perp} - g_f| .$$

The renormalization depends on the relative importance of $u = g_c^2$ and $(g_f)_{\text{DD}}$. The DD would lead to $g_{\parallel} \rightarrow -\infty$, whereas SOC would lead to $g_{\parallel} \rightarrow +\infty$ [notice, therefore, that a numerical study of the equations shows that $g_{\parallel} \rightarrow -\infty$ needs $u \gg (g_f)_{\text{DD}}$, i.e., a strong SOC].

For $g_c = 0$ with small modifications¹² one can see that the spin part of our Hamiltonian (11) gives the partition function of a two-dimensional Coulomb gas of charges and magnetic monopoles. This model is related to the two-dimensional eight-vertex and Ashkin-Teller models, as well as to the one-dimensional XYZ spin chain.¹³⁻¹⁵

We have demonstrated here that spin-orbit coupling or electron-electron dipolar interactions lead to spin-dependent and -nonconserving electron-electron interactions. Their main effect is the opening of a gap in the spin-excitation spectrum in the whole phase diagram, except on the critical plane. This leads to completely anisotropic ($x \neq y \neq z$) spin-density-wave and triplet-superconducting phases and to an anisotropic spin susceptibility. The importance of these effects is governed by the spin-excitation gap Δ_σ . In a real, quasi-one-dimensional system, generally there are phase transitions with a nonzero critical temperature T_c , due to the existence of some kind of interchain coupling. If $T_c < \Delta_\sigma$, we would expect, due to the gap in the spin-excitation spectrum, an anisotropic, thermally activated spin susceptibility, as well as a strongly anisotropic ordered phase below T_c . On the other hand, we note that for $T \gg \Delta_\sigma$ the exponents are reduced:

$$\alpha_i(T \gg \Delta_\sigma) = \alpha_i(T \ll \Delta_\sigma) - 1 . \quad (18)$$

Consequently, only below Δ_σ the correlation functions diverge rapidly. A not too weak three-dimensional coupling then would lead to a phase transition at a temperature close to Δ_σ , so that the effects of a nonzero Δ_σ would only be observable over a narrow temperature range.

We try now to give some qualitative estimate of the importance of the effects discussed here in real quasi-one-dimensional compounds like $(\text{TMTTF})_2\text{X}$ or $(\text{TMTSF})_2\text{X}$. Although we are interested in temperatures lower than the transverse hopping matrix element t_\perp this one-dimensional model may still be valid because, as pointed out before,⁷ due to one-dimensional correlations the one- to three-dimensional crossover temperature can be much smaller than t_\perp . In those compounds a thermally activated spin susceptibility is not observed^{2,4} and anisotropy only exists in a narrow temperature range above the phase transition into the antiferromagnetic (SDW) state. From the above we then have an upper limit for Δ_σ :

$$\Delta_\sigma \leq T_c \approx 10-20 \text{ K} . \quad (19)$$

It has been argued⁶ that for $(\text{TMTTF})_2\text{X}$ and for $(\text{TMTSF})_2\text{X}$ compounds one has $g_1 > 0$. Consequently, in our (simplified) model SOC alone would lead to a SDW_z fluctuation, whereas the DD interactions give a dominant SDW_y divergence, in agreement with experiment (our y axis is roughly the crystalline b axis). Thus dipolar interactions may possibly play a major role in determining the magnetic anisotropies [notice that in the renormalization equation (12) the SOC contribution in g_f and g_1 nearly cancel them-

selves to lowest order [cf. Eq. (5)]. This has been pointed out previously,⁴ based on the observation that the magnetic anisotropy of $(\text{TMTTF})_2\text{X}$ and $(\text{TMTSF})_2\text{X}$ are very similar, whereas SOC is much weaker in sulfur [$(\text{TMTTF})_2\text{X}$] than in selenium [$(\text{TMTSF})_2\text{X}$] compounds.

It is beyond the scope of the present paper to do molecular calculations of spin-orbit coupling effects. We rather try to obtain a semiquantitative estimate. The results of Ref. 16 and a Hartree-Fock calculation for Se atoms¹⁷ show that the SOC is of order of 5% of the bandwidth in $(\text{TMTSF})_2\text{X}$ compounds. The prefactor in the matrix elements [Eq. (6)] contains the long-range part of the three-dimensional Coulomb interaction, where screening by adjacent chains has to be included,¹⁸ and for $(\text{TMTSF})_2\text{X}$ this prefactor is of order of the bandwidth. So the SOC energies are some 10^{-2} of a bandwidth. As the diagrams of Fig. 1 depend on the SOC squared the overall values of $g_{111} - g_{112}$ and g_f are probably of the order of 1 K, but an order of magnitude smaller in the sulfur analogue compounds $(\text{TMTTF})_2\text{X}$ due to the much weaker SOC. The magnitude of the DD term is hard to estimate: Although it is usually weaker than the SOC in the DD interactions the electron-electron distance intervenes, which can be smaller than the atom-electron

one. From these numerical estimates we expect Δ_σ to be considerably smaller than the above upper limit 10–20 K. A small Δ_σ can also be deduced from the small spin-flop field H_{sf} . A simple argument gives $g\mu_B H_{\text{sf}} \approx \Delta_\sigma$, and the experimental values of H_{sf} then imply $\Delta_\sigma \approx 0.5$ K. Consequently, we conclude that spin anisotropies play no major role in the one-dimensional regime of $(\text{TMTTF})_2\text{X}$ and $(\text{TMTSF})_2\text{X}$ compounds. The anisotropies observed above T_c are probably due to three-dimensional fluctuation effects. In that case, the much larger coherence volume can considerably enhance anisotropies. However, we expect the microscopic origin of anisotropy to be the same both in the one- and three-dimensional regime.

We finally remark that there are large regions of coexistence SDW and TS fluctuations in our phase diagram (Fig. 2). The possible importance of coexisting supraconducting and SDW fluctuations in $(\text{TMTSF})_2\text{X}$ compounds has been pointed out before;^{1,7} however, the question of the possible triplet character of these fluctuations is still open.

A detailed version of the present paper will be published subsequently.

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