On the origin of the anomalous upper critical field in quasi–one-dimensional superconductors

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Abstract – The upper critical field, \(H_{c2}\), in quasi-1D superconductors is investigated by the weak coupling renormalization group technique. It is shown that \(H_{c2}\) greatly exceeds not only the Pauli limit, but also the conventional paramagnetic limit of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state. This increase is mainly due to the quasi-1D fluctuations effect as triggered by interference between unconventional superconductivity and density-wave instabilities. Our results give a novel viewpoint on the large \(H_{c2}\) observed in TMTSF-salts in terms of a \(d\)-wave FFLO state that is predicted to be verified by \(H_{c2}\) measurements under pressure.

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Introduction. – Thirty years after the discovery of organic superconductivity in the quasi–one-dimensional (quasi-1D) molecular conductor (TMTSF)\textsubscript{2}PF\textsubscript{6} [1], the nature of this phase has not been entirely clarified in spite of intensive experimental and theoretical efforts devoted to this end. An enduring problem concerns the properties of superconductivity under magnetic field, in particular the issue of the upper critical field \(H_{c2}\) in these materials, which is the topic of the present letter. The value of \(H_{c2}\) determined, for instance, by resistivity measurements, drastically exceeds the so-called Clogston or Pauli limit, \(H_P\). The excess occurs when the magnetic field is oriented parallel to the plane of (TMTSF)\textsubscript{2}X (\(X = \text{PF}_6\) [2], ClO\textsubscript{4} [3]), that is when the pair breaking orbital effect is virtually quenched by anisotropy. There are basically two possibilities that have been put forward to explain the excess of \(H_{c2}\): the existence of a spin-triplet pairing or the presence of a Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) phase.

The constant spin susceptibility, \(\chi\), as obtained from the first Knight shift measurements across \(T_c\) for \(X = \text{PF}_6\) under pressure gave support to the former scenario for triplet superconductivity [4]. On the other hand, more recent NMR results on the ambient pressure superconductor \(X = \text{ClO}_4\), display a decreasing \(\chi\) below \(T_c\), at least in low fields, which is compatible with singlet rather than triplet pairing [5]. As for the FFLO state, an estimation of the paramagnetic limit, \(H_{c2}^{\text{FFLO}}\), puts it lower than the \(H_{c2}\) observed in (TMTSF)\textsubscript{2}X [6], which would make this scenario unlikely on a theoretical basis. However, recent detailed resistivity measurements on the \(X = \text{ClO}_4\) compound using a rotating field, display the signature of non-uniform superconductivity at high fields, suggesting that a FFLO state could be realized [7,8]. The amplitude of \(H_{c2}\) determined by the bulk measurements such as the nuclear relaxation rate [5], magneto-torque [9] and specific heat [10], is lower than that of resistivity measurements, indicating the existence of a high field interval \(H_{c2}^{\text{bulk}} < H < H_{c2}^{\text{resist}}\), which we term as a “transient region”, where incomplete superconductivity is realized.

On the theoretical side, the possibility of triplet superconductivity in an interacting quasi-1D electron system has been investigated on a microscopic basis in various situations. In zero field, \(f\)-wave triplet pairing has been shown to compete with \(d\)-wave singlet pairing, when long-range intrachain interactions become sufficiently large [11–14]. The competition was also shown to emerge when interchain repulsive interactions are finite, albeit small [15,16]: or as the amplitude of the on-site interaction is huge [17]. A field-induced singlet to triplet
pairing crossover has been also proposed to occur on phenomenological grounds [12,14,18]. A similar transition under field has been shown to take place microscopically using the mean-field theory [19], or when in a similar framework, long-range interactions are included [20–22]. The $H$-$T$ phase diagram thus obtained, however, departs from observation obtained for the bulk [7,8,10]; it would also give a temperature-independent spin susceptibility in the metallic state, at variance with experiments [23,24].

The FFLO state in the quasi-1D geometry has been examined in great detail in the framework of the BCS mean-field theory, which predicts its existence in the region of small $T_c$ [25–27]. However, the mean-field theory neglects the influence of fluctuations, in particular, those linked to many-body processes resulting from interference between superconducting (SC) and density-wave (DW) pairings. Such a mix of pairing mechanisms is present at every order of perturbation theory for quasi-1D interacting electron systems and is responsible for the dynamical generation of $d$-wave superconducting pairing from spin fluctuations [15,24,28,29]. For compounds like (TMTSF)2X, where superconductivity is just found in the close proximity of a SDW instability, the interference is expected to play an important role. Fully taken into account by the renormalization group method (RG), the interference proved to have a non-trivial impact not only on the mechanism and nature of the superconductivity [14–16,30], but also on the property of the metallic state above $T_c$. It carries over well below the crossover temperature scale for the onset of transverse coherent single-electron motion where marked deviations from an ordinary Fermi liquid can be found [24,29,31,32]. The repercussions of these fluctuation effects on the stability of superconductivity under field are until now unexplored.

In this letter we tackle the problem of the upper critical field in quasi-1D superconductors close to a spin density-wave instability and clarify the influence of quasi-1D fluctuations on superconductivity under magnetic field. For this purpose, we extend the renormalization group (N-chains RG) technique [24] to incorporate a finite Zeeman splitting, and study the influence of interference on $d$-wave singlet superconductivity under magnetic field. It is shown that the $H_{c2}$ exceeds not only $H_F$, but also the mean-field prediction of $H_{c2}^{\text{FFLO}}$ to a great extent. The predicted non-universal character of the effect, as a function of interactions and nesting alterations of the Fermi surface, correlates the degree of the interference between SC and DW. This gives the possibility to confirm the relevance of the present theory by new experiments under pressure and field.

Theory. – We consider a quasi-1D model of a square array of weakly coupling metallic chains, taken away from half-filling for simplicity. The hopping integrals $t_i$ obey the following anisotropic sequence: $t_a \gg t_b \gg t_c$ along the $a$-, $b$-, and c-axis. The magnetic field is oriented such that its impact is restricted to the Zeeman splitting alone. Such conditions can be realized in practice when the field is oriented in the $(a, b)$-plane, which easily suppresses the orbital motion along the least conducting $c$-axis at small enough $t_c$ [25]. The remaining relevant electron spectrum in the remaining $(a, b)$-plane is given by

$$E_{\pm}(\mathbf{k}) = v_F(\pm k_a - k_F) + \xi_b(k_b) + \mu_B \sigma H,$$

where $\mu_B$ is the Bohr magneton (in the following, we use $H = \mu_B H$ and $H = 1$, and $\xi_b(k_b) = -2i(k_b \cos k_b - 2i \cos 2k_b)$. Here the longitudinal part of the spectrum has been linearized around $\pm k_F$, with the longitudinal Fermi velocity $v_F = 2t_a \sin k_F$. The next-nearest interchain hopping, $t_b'$, breaks the perfect nesting condition of the Fermi surface (FS).

Electron-electron scattering matrix elements are expressed in terms of interactions between electrons on opposite Fermi sheets [33], namely as $g_1$ and $g_1'$ for backward scattering with parallel and antiparallel spins, and $g_2$ for forward scattering. For the Hubbard model, we have $g_1 = 0$. $g_1' = U/\pi v_F \equiv U$, as bare normalized couplings, where $U$ is the on-site repulsion of the Hubbard model. We employ a momentum shell RG approach that is essentially equivalent to the one-particle irreducible scheme at one-loop level [16,24,34,35].

The one-loop flow equations so obtained are

$$\frac{\partial g_{||}}{\partial t} = \frac{g_{||}}{6} \left( I_{C}^0 - I_{F}^0 \right) - g_{11}g_{11} I_{F}^4,$$

$$\frac{\partial g_{11}}{\partial t} = -(g_{11} g_{22} + g_{22} g_{11}) \left( I_{C}^0 + I_{F}^4 \right)/2 - (g_{11} g_{||} + g_{||} g_{11}) \left( I_{F}^0 + I_{F}^4 \right)/2,$$

$$\frac{\partial g_{22}}{\partial t} = g_{22} g_{22} \left( I_{F}^0 - I_{C}^0 \right) - g_{11}g_{11} I_{F}^4,$$

where the momentum dependences of the couplings on the left-hand sides of eqs. (2)–(4) are $g_1(k_{1b}, k_{2b}, k_{3b})$ with incoming $k_{1b}$, $-k_2$, and outgoing $k_3$ and $-k_4 = k_1 - k_3 - k_4$. On the right-hand side, the momentum dependence of coupling products are in order: $g(k_{1b}, k_{2b}, k_{3b}, k_{4b})g(k_1, k_2, k_3, k_4)$ for the Cooper channel (particle-particle channel: SC fluctuations) that involves $I_{C}^0$ and $g(k_{1b}, k_{2b}, k_{3b}, k_{4b})$ for the Peierls channel (particle-hole channel: DW fluctuations) that involves $I_{F}^0$. The summations with respect to $k_{3b}$ are taken as $\sum_{k_{3b}}$, where $N$ is the number of chains in the $(a, b)$-plane. The loop shell integration yields

$$I_{\nu \ell}^0 = \frac{E_{\ell}^4}{4} \sum_{\lambda = -1}^{1} \left( \frac{\Theta(|E_\nu + 2 + \lambda A_{\nu}^{ab} - E_\ell/2)| - E_\ell/2}{E_\ell + \lambda A_{\nu}^{ab}} \right) \times \left\{ \tanh \frac{E_\nu}{4 T} + \tanh \frac{E_\nu/2 + \lambda A_{\nu}^{ab}}{2 T} \right\},$$

where

$$A_{\nu}^{ab} = \xi_a(k_a) - \xi_b(q_b - k_b) - \xi_a(k_{1b}) + \xi_b(k_{2b}) - \alpha h,$$

$$A_{\nu}^{bh} = \xi_a(k_a) + \xi_b(k_b - Q_b) - \xi_a(k_{3b}) - \xi_b(k_{3b}) - \alpha h,$$

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and $\Theta(x) = 1, 1/2$ and 0 for $x > 0$, $x = 0$ and $x < 0$, respectively. The scaled bandwidth is $E_x = E_0 e^{-\xi}$ at the RG step $\ell > 0$. The initial band width $E_0 (= 2v_F k_F)$ is taken as unity. The magnetic field acts as a cutoff for certain logarithmic singularities through $\Lambda^{\phi}_{\ell}$ in the Cooper and Peierls channels, in a way similar to nesting deviations $t'_b$ for the Peierls singularity.

From refs. [24,34], the dimensionless response function for singlet-type pairing at $q = 0$, reads

$$\chi^{e,o}_{\text{ss}}(T) = \frac{2}{\pi v_F} \int_0^\infty d\ell \left\{ z^{e,o}_{\text{ss},n}(\ell) \right\}^2 I^{2h}_C(q_b = 0), \tag{8}$$

where the pair vertex obeys the flow equation

$$\partial_{\ell} z^{e,o}_{\text{ss},n} = - \frac{1}{2} z^{e,o}_{\text{ss},n} ((g_{1\perp} + g_{2\perp}) I^{2h}_{C}(q_b = 0)). \tag{9}$$

In eq. (8), $I^{2h}_{C}$ is independent of $k_b$’s, which is obvious from eq. (6) with $q_b = 0$ and $k_b = k_b$. In eq. (9), the momentum dependences for the couplings and loop are $g_{1\ell} (k_b, k_b - q_b, k_b')$ and $I^{2h}_{C}(q_b, k_b')$. $\{ \ldots \}^{e,o}$ denotes the even- (odd)-$n$-th Fourier component as $\langle f(k_b, k'_{b'})^{e,o}(\ell) \rangle = N^{-2} \sum_{k_b,k'_{b'}} f(k_b, k'_{b'}) (\cos\sin n) k_b (\cos\sin n) k'_{b'}$. $z^{e,o}_{\text{ss},n}$ is the pair vertex for the singlet superconductivity: $z^{e,o}_{\text{ss},0}$ is for the $s$-wave, and $z^{e,o}_{\perp 1}$ is for the $d$-wave. In the case of triplet pairings, $z^{e,o}_{\perp 0}$ is for the $p$-wave, and $z^{e,o}_{\perp 1}$ is for the $f$-wave. Note that the superscripts “$e$” and “$o$” denote the even-and odd-function part of the “$k_b$-dependence” of the gap function, so that $z_{n}$ is not necessarily an odd-function with respect to $k_b$ even for the triplet pairings [14]; the requirement of odd-parity symmetry for the triplet pairing is already satisfied by the combination of the couplings in the pair-flow equation, such as $g_{2\perp} - g_{1\perp}$.

For the FFLO-type pairing, $q \neq 0$, and the formulation is modified due to the mixture of singlet and triplet pairing as follows:

$$\chi^{e,o}_{\text{test}}(T) = \frac{1}{\pi v_F} \int_0^\infty d\ell \left\{ \left( z^{e,o}_{\text{test},n}(\ell) \right) \right\}^2 I^0_{C}(q_b = 0) + \left\{ \left( z^{e,o}_{\text{test},n}(\ell) \right) \right\}^2 I^{2h}_{C}(q_b = 0), \tag{10}$$

where the pair vertex is renormalized as

$$\partial_{\ell} z^{e,o}_{\text{test},n} = - \frac{1}{2} \left( z^{e,o}_{\text{test},n} g_{1\perp} I^0_{C} + z^{e,o}_{\text{test},n} g_{1\perp} I^{2h}_{C} \right), \tag{11}$$

$$\partial_{\ell} z^{e,o}_{\text{test},n} = - \frac{1}{2} \left( z^{e,o}_{\text{test},n} g_{1\perp} I^0_{C} + z^{e,o}_{\text{test},n} g_{1\perp} I^{2h}_{C} \right). \tag{12}$$

Here, $z^{e,o}_{\text{test}}$ is the pair vertex for a Cooper pair of electrons with $\uparrow \downarrow$ ($\uparrow \uparrow$) spins and center-of-mass momentum $q$, which is determined by the shift of the Fermi surface due to the magnetic field as $q = (2h/eF, q_b)$. Similar definitions hold for the other response functions [24]. For the determination of the strongest singularity corresponding to the dominant instability, we compare the derivatives of the response functions, $\partial \chi_{\mu}$. In the following, we set $t_b = 0.1 E_0$, $t'_b = 0.01 E_0$, $g_{||} = 0.0$ and finite repulsive $\tilde{U}$.

Fig. 1: (Colour on-line) Response functions, $\partial \chi_{\mu}(h)$, as a function of magnetic field for $\tilde{U} = 1.0$, $T = 10^{-3} E_0$ and $N = 31$. The subscript of $p$ and $f$ denotes the $s$, $t$ of triplet pairing; $s_t = 1$ is the equal spin pairing and $s_t = 1$ is the triplet pairing with antiparallel spins. SDW and $\text{SDW}_{xy}$ are the longitudinal and the transverse spin density wave, respectively.

**Results.**

Response functions. The magnetic-field dependences of the derivative of the response functions, $\partial \chi_{\mu}(H)$, for $N = 31$ chains are shown in fig. 1. Here we set $g_{1\perp} = g_{2\perp} = 1.0$ ($\tilde{U} = 1.0$), and $T = 10^{-3} E_0$. The SDW correlations are reduced (compared to the $d$-wave) by nesting deviations due to $t'_b$. At low fields, the most dominant instability is for $d$-wave pairing with $q = 0$. (Hereafter we call the non–FFLO-type ($q = 0$) $d$-wave pairing just as “$d$-wave pairing”.) It is reduced for $h \gtrsim 10^{-3} E_0$, reflecting the cutoff effect of $h$. The instability that closely follows is towards the $d$-wave FFLO (charged FFLO) order, which is also reduced for $h \gtrsim T$, albeit less than $d$-wave, so that it dominates at high field. In the limit of very high field, $h \gtrsim 10^{-2} E_0$, the $d$FFLO and the transverse SDW (SDW$_{xy}$) are prevalent with increasing $H$, feeding one another through the coupling $g_{2\perp}$ as a result of interference.

H-T phase diagram. Next, we show the results of the temperature dependence of $H_{c2}$ for $\tilde{U} = 0.6, 0.8$, and 1.0 in fig. 2. Significant features are obtained. First the trace of $H_{c2}(T)$, which is determined by the singularity in $\partial \chi_{\mu}$, greatly exceeds not only the Pauli limit, $H_{P} \approx 1.25 t_c$, but also the paramagnetic limit of the FFLO state, $H_{c2}^{\text{FFLO}}$ in 2D ($\simeq 1.78 t_c$) and 3D ($\simeq 1.34 t_c$) isotropic cases [36]. Significantly enough, the limiting field ratio $H_{c2}^{\text{FFLO}} / t_c$ found here is non-universal and depends on the strength of interactions.

The origin of such an increase is primarily due to quasi-1D fluctuations introduced by SC-DW interference. The mixing between the Peierls and Cooper scattering channels in eq. (3) induces an attractive coupling for $d$-wave superconductivity from spin fluctuations, which is scale dependent all the way down to the lowest energy scale.

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of the flow, that is \( T_c \). This contrasts with the standard single-channel or mean-field approach to superconductivity for which the attractive coupling that feeds the Cooper pairing channel is fixed and tied to the high-energy cutoff of the theory. This can be further checked by comparing the RG results with those of the RPA — ladder diagrammatic summation — limit, which is obtained by switching off the Peierls contribution to the flow of coupling constants in eqs. (2)–(4) \((P^b_{\alpha h} = 0)\). There will be then no interference and by assuming an attractive coupling \( \tilde{U} < 0 \), an \( s \)-wave SC instability will thus be realized in the mean-field way, while keeping the same geometric condition on the FS.

The results of ladder summation are also shown in fig. 2 (the bottom solid curve). In this case \( H_{c2}(T) \) almost agrees with the ordinary one \([36]\), \( H_{c2}(T) \approx H_P \sqrt{1 - T/T_c} \) around \( T_c \), and extrapolates also towards \( H_P \) at zero temperature. As for the FFLO state in the ladder limit, \( H_{c2}^{FFLO}(T) \) is higher than the isotropic value \( H_{c2,2D}^{FFLO} \). This slight increase of \( H_{c2}^{FFLO}(T) \) is due to a geometric property of the quasi-1D FS. In general, there is a geometric restriction to finite momentum FFLO pairing, as shown in fig. 3(a). This restriction is partly removed when the FS shows nesting (fig. 3(b)). In the case of a quasi-1D FS, however, there is no restriction and FFLO pairing is enhanced, as shown in fig. 3(c). It should be noted that for the ladder approximation, while the amplitude of \( \tilde{U} \) does affect both \( T_c \) and \( H_{c2} \), the traces given by the ratio \( H_{c2}/T_c \) versus \( T/T_c \) are essentially invariant and scale to the same universal curve shown in fig. 2 for the whole range of small attractive couplings, at variance with the non-universality displayed by the full one-loop RG results.

**Discussion.** — Let us now discuss the implications of the present results for \((TMTSF)_{2}X\) compounds. For \((TMTSF)_{2}ClO_4\), the specific-heat measurements of the bulk critical field, \( H_{c2}^{bulk} \), along both \( a \) and \( b' \) directions agree with the Pauli limit at low temperature, but \( H_{c2}^{bulk} \) is significantly lower than \( H_{c2}^{nest} \), as extracted from the resistivity onset. This allows to define a rather large “transient region”, \( H_{c2}^{bulk} < H < H_{c2}^{nest} \), in the \((H, T)\)-plane where incomplete superconductivity is found. While there are no specific-heat data available for \((TMTSF)_{2}PF_6\) under field, the values of \( H_{c2}^{nest} \) as obtained close to the critical pressure are similar to those of \((TMTSF)_{2}ClO_4\) at ambient pressure. This suggests that the transient region for both compounds is of similar size \([2]\).

The transient region observed experimentally should correspond to the \( d\)FFLO region of the above two-dimensional theory (fig. 2). It seems to even exceed in size the one found experimentally at intermediate initial coupling \( \tilde{U} \). However, \( d\)FFLO superconductivity is expected to be quite sensitive to finite impurity scattering in these materials known to be more pronounced for \((TMTSF)_{2}PF_6\) at ambient pressure. This suggests that the transient region for both compounds is of similar size \([2]\).

Finally, a crucial test of the present theory of the \( d\)FFLO state in materials like the Bechgaard salts would be clearly the comparison of the \( H-T \) phase diagram at various pressures, as can be measured, for instance, by resistivity. In the present model the spin fluctuation, which induces attractive coupling for \( d \)-wave superconductivity, is known to be strongly pressure dependent via the anti-nesting parameter \( t'_b \), which besides \( \tilde{U} \) simulates pressure effects in the model \([16, 29]\). Actually, the ratio \( H_{c2}/T_c \) decreases with increasing \( t'_b \) as shown in fig. 4, namely, the size of the transient region will shrink in size as pressure increases. The pressure-dependent, non-universal, reduction of \( H_{c2} \) is a direct consequence of non-universality linked to the interference between SDW and Cooper pairing.
Summary. – In the present work, $H_c2$ for the quasi-1D superconductivity has been calculated for the Hubbard model by the newly developed N-chain RG under magnetic field. We have shown that when constructive interference between density-wave and Cooper pairings is fully taken into account, the $H_c2$ of a quasi-1D d-wave superconductor greatly exceeds not only the Pauli limit, but also the conventional paramagnetic limit of the FFLO state. In a material like (TMTSF)$_2$ClO$_4$, the results presented here propose a new way for the reconciliation of transport [7,8], magneto-torque [9], specific-heat [10] and NMR data [5]. The anomalous $H_c2$ enhancement found from this mechanism proves to be non-universal in both interaction and anti-nesting strengths, allowing the possibility for a direct experimental test for the predicted correlation between the $H_c2$ excess and $T_c$ under pressure.

In conclusion, we have proposed a mechanism of the anomalous broadening of the upper-critical-field region as seen from transport measurements in quasi-1D organic superconductors like the Bechgaard salts series. The mechanism proposed is a direct consequence of the interplay between magnetism and unconventional superconductivity in these materials under magnetic field.

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