Possible restoration of superconductivity in the quasi-one-dimensional conductor Li\(_{0.9}\)Mo\(_{6}\)O\(_{17}\) in feasibly high pulsed magnetic fields, \(H \approx 100\) T.

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We present a theoretical study of restoration of superconductivity in the form of the triplet reentrant superconducting phase in a quasi-one-dimensional (Q1D) conductor. Substitution of known band and superconducting parameters of the presumably triplet Q1D superconductor Li\(_{0.9}\)Mo\(_{6}\)O\(_{17}\) into our theoretical equations shows that such restoration can happen in feasibly high non-destructive pulsed magnetic field of the order of \(H \approx 100\) T. We investigate in detail how small inclinations of a direction of a magnetic field from its best experimental geometry decrease superconducting transition temperature of the reentrant phase, which is important for its possible experimental discovery.

PACS numbers: 74.20.Rp, 74.70.Kn, 74.25.Op

I. INTRODUCTION

Physical properties of quasi-one-dimensional (Q1D) superconductors have been intensively studied since the discovery of superconductivity in the Bechgaard salts - superconductors in the chemical family \((\text{TMTSF})_2X\), where \(\text{X} = \text{ClO}_4, \text{PF}_6\), etc [1,2]. Early experiments alluded to the unconventional nature of these superconductors, as non-magnetic impurities destroyed superconductivity [3,4] and the so-called Hebel-Slichter peak in the NMR data was not observed [5]. Recent experiments [6] have firmly supported these early findings. Despite 30 years of intensive investigations, the nature of superconductivity in the Bechgaard salts is still controversial. On one hand it was shown [7-9] that superconducting phase in \((\text{TMTSF})_2\text{ClO}_4\) compound is more likely of \(d\)-wave type with zeros on Q1D Fermi surface. On the other hand, in \((\text{TMTSF})_2\text{PF}_6\) compound there is still some chance of a spin triplet superconducting pairing [10-12].

Triplet superconductors with layered Q1D Fermi surfaces (FS) were theoretically shown to exhibit very unusual magnetic properties in a high magnetic field perpendicular to the chains and parallel to the layers. More precisely, it was demonstrated [13-16] that superconductivity can be restored as a pure two-dimensional phase when the magnetic field is high enough to localize conducting electrons on Q2D layers. This happens when the typical ”sizes” of electron trajectories become less than the inter-plane distance due to the so-called quantum \(3D \rightarrow 2D\) dimensional crossover [2]. (Note that this phenomenon, which we call reentrant superconductivity, was first suggested in Ref.[13] and is different from the quantum limit (QL) superconductivity, suggested for a pure 3D case by Tesanovic and Razolt [17] and for a Q1D case by us [18].)

In this context, the possible triplet superconductor Li\(_{0.9}\)Mo\(_{6}\)O\(_{17}\) presents unique opportunity to study superconductivity in high magnetic fields. Li\(_{0.9}\)Mo\(_{6}\)O\(_{17}\) is a Q1D, layered transition metal oxide, structurally similar to the Bechgaard salts. It is metallic at high temperatures and undergoes a superconducting transition at \(T_c \approx 2.2\) K. Recently, Mercure et al. [19] have shown that superconductivity in the Li\(_{0.9}\)Mo\(_{6}\)O\(_{17}\) compound exceeds the so-called Clogston-Chandrasekhar paramagnetic limit [20] by 5 times. Our theoretical analysis [21,22] of the experimental curves in Ref. [19] showed excellent quantitative agreement between the experimental properties and the theoretical predictions for a spin-triplet superconductor in the absence of Pauli destructive effects. Thus, the presumably triplet superconductor Li\(_{0.9}\)Mo\(_{6}\)O\(_{17}\) is a major candidate for experimental discovery of reentrant superconductivity. We note that the possibility of existence of reentrant superconductivity phenomenon is restricted by such triplet phases where the spin-splitting Pauli effects against superconductivity are absent. The band and superconducting parameters of this compound have been calculated in Ref. [21], along with the most likely triplet pairing scenario presented in Refs. [21,22]. The latter shows that a nodeless scenario of triplet superconductivity parametrized by an order parameter that changes its sign on the two sheets of the Q1D Fermi surface leads to the experimentally observed destruction of superconductivity by pure orbital effects in a magnetic field parallel to conducting chains.

The goal of this paper is twofold. First, we suggest experimental discovery of the reentrant superconducting phase in the possible spin-triplet, Q1D layered superconductor Li\(_{0.9}\)Mo\(_{6}\)O\(_{17}\) using the currently experimentally available pulsed (non-destructive) magnetic fields of order \(H \approx 100\) Tesla. Our calculations show that such fields, parallel to the layers and perpendicular to the conducting chains lead to the appearance of the reentrant superconductivity phenomenon in Li\(_{0.9}\)Mo\(_{6}\)O\(_{17}\) below a reentrant transition temperature of \(T^*(H = 100T) \approx T_c/2 \approx 1\)K. Second, we calculate the angular dependence of \(T^*(\alpha, H)\) to account for inclination of the same magnetic field from the optimal geometry of the corresponding experiment. The latter will allow to conduct the experiments with necessary accuracy.
II. REENTRANT SUPERCONDUCTING PHASE IN \(\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}\)

For our present theoretical analysis of the reentrant phase, we initially consider a Q1D conductor in a magnetic field parallel to the layers and perpendicular to the conducting chains, as shown in Fig.1. The Q1D Fermi surface, shown in Fig.2, consists of two open, slightly warped sheets centered at \(p_x = \pm p_F\). The anisotropic electron spectrum, with \(t_x \gg t_y \gg t_z\) can be linearized on the right (+) and left (-) sheets as follows:

\[
e^{\pm}(\vec{p}) = \pm v_F(p_x - p_F) - 2t_y \cos(p_ya_y) - 2t_z \cos(p_za_z). \tag{1}
\]

The magnetic field and the vector potential have the following components:

\[
\vec{H} = (0, H, 0), \quad \vec{A} = (0, 0, -Hx). \tag{2}
\]

The Hamiltonian is obtained from Eq.(1) by the Peierls’ substitution method [2,23]:

\[
(p_x - p_F) \rightarrow -i \frac{d}{dx}, \quad p_ia_i \rightarrow p_ia_i - \frac{ea_i}{c}A_i,
\]

Motion parallel to the magnetic field (along the \(y\) direction) does not lead to twisting of electron orbits, allowing us to consider the following reduced Hamiltonian for a system in a magnetic field:

\[
\hat{H}^\pm = \mp iv_F \frac{d}{dx} - 2t_z \cos \left(p_za_z + \frac{\omega_z}{v_F}x\right), \tag{3}
\]

where the transverse oscillation frequency is

\[
\omega_z = \frac{ev_Fa_zH}{c}. \tag{4}
\]

The Schrödinger-like equation, \(\hat{H}\Psi_\epsilon = \epsilon\Psi_\epsilon\), for the Q1D wave functions becomes

\[
\left[\mp iv_F \frac{d}{dx} - 2t_z \cos \left(p_za_z + \frac{\omega_z}{v_F}x\right)\right] \Psi_\epsilon^\pm(x; p_z) = \epsilon\Psi_\epsilon^\pm(x; p_z), \tag{5}
\]

where the energy \(\epsilon\) is measured with respect to the Fermi energy. By direct substitution into Eq.(5) we can check that the exact solutions for the Q1D electron wave functions are

\[
\Psi_\epsilon^\pm(x; p_z) = \frac{1}{\sqrt{2\pi v_F}} \exp \left(\pm \frac{i\epsilon}{v_F}x\right) \times \exp \left\{\mp \frac{2it_z}{\omega_z} \left[\sin \left(p_za_z + \frac{\omega_z}{v_F}x\right) - \sin(p_za_z)\right]\right\}. \tag{6}
\]

The normalization of these wavefunctions is such that

\[
\int_{-\infty}^{\infty} \Psi_\epsilon^\pm(x; p_z)\Psi_{\epsilon'}^\pm(x; p_z)' \, dx = \delta(\epsilon - \epsilon').
\]
Having obtained the wave functions, we construct the finite temperature Green’s functions according to the standard procedure [24]:

\[ G_{i\omega_n}^\pm (x, x'; p_z) = \sum_x \frac{\Psi_{i\omega_n}^\pm (x, p_z) \Psi_{i\omega_n}^\pm (x', p_z)^*}{i\omega_n - \epsilon}, \quad (7) \]

where the Matsubara frequency for fermions is \( \omega_n = \pi T (2n + 1) \). Performing the sum in Eq.(7) we obtain the expressions for the Green’s functions:

\[ G_{i\omega_n}^\pm (x, x'; p_z) = -\frac{i}{v_F} \text{sgn}(\omega_n) e^{\pm \omega_n (x-x')/v_F} g^\pm (x, x'; p_z), \quad (8) \]

where

\[ g^\pm (x, x'; p_z) = \exp \left\{ \frac{2i\epsilon}{\omega_z} \left[ \sin \left( p_z a_z + \frac{\omega_z}{v_F} x \right) - \sin \left( p_z a_z + \frac{\omega_z}{v_F} x' \right) \right] \right\}. \quad (9) \]

The equation to determine superconducting order parameter is obtained from Gor’kov’s equations for non-uniform superconductivity [24]. To this end, we take the simplest spin triplet order parameter that changes its sign on the two sheets of the Q1D Fermi surface. Note that it satisfies experimental data [19] and is insensitive to Pauli spin-splitting effects,

\[ \hat{\Delta} = \hat{I} \text{sgn}(p_z) \Delta(x), \quad (10) \]

where \( \hat{I} \) is a unit matrix in spin space, and \( \text{sgn}(\pm p_F) = \pm 1 \), while \( \Delta(x) \) is the solution of the so-called gap integral equation

\[ \Delta(x) = g \int K(x, x') \Delta(x') dx', \quad (11) \]

where \( g \) is the effective electron coupling constant, and the kernel is

\[ K(x, x') = T \sum_{\omega_n} \frac{g^+ (x, x'; p_z) g^- (x', -p_z)}{i\omega_n - \epsilon} \quad (12) \]

The products of Green’s functions in Eq.(12) are averaged over the momentum component \( p_z \). By using the expressions for the Green’s functions in Eq.(9) it can be shown that

\[ \left\langle g^+ (x, x'; p_z) g^- (x, x'; -p_z) \right\rangle_{p_z} = \exp \left\{ \frac{8i\epsilon}{\omega_z} \left[ \sin \left( \frac{\omega_z}{2v_F} (x - x') \right) - \sin \left( \frac{\omega_z}{2v_F} (x + x') \right) \right] \right\} \]

\[ J_0 \left\{ \frac{8i\epsilon}{\omega_z} \left[ \sin \left( \frac{\omega_z}{2v_F} (x - x') \right) - \sin \left( \frac{\omega_z}{2v_F} (x + x') \right) \right] \right\}, \quad (13) \]

where \( J_0 \) is the Bessel function. The summation over both the positive and the negative Matsubara frequencies in Eq.(12) results in a factor

\[ \sum_{\omega_n} e^{-2\omega_n (x-x')/v_F} = \frac{1}{2 \sinh \left( \frac{2\pi T}{\omega_z} |x - x'| \right)} \quad (14) \]

Substituting the results of Eqs.(13),(14) into the definition of the kernel in Eq.(12), we obtain the gap integral equation for the superconducting order parameter [13]:

\[ \Delta(x) = \frac{g}{2} \int_{|x-x'|>d} \frac{2\pi T \Delta(x') dx'}{v_F \sinh \left( \frac{2\pi T}{\omega_z} |x - x'| \right)} \times \]

\[ J_0 \left\{ \frac{8i\epsilon}{\omega_z} \left[ \sin \left( \frac{\omega_z}{2v_F} (x - x') \right) - \sin \left( \frac{\omega_z}{2v_F} (x + x') \right) \right] \right\}, \quad (15) \]

where \( d \sim v_F/\Omega \) is the cutoff distance. Note that the equation obtained above is quite general, and provides various descriptions of superconductivity for Q1D layered compounds. For example, it can be shown that for temperatures \( T \approx T_c \) and low enough magnetic fields Eq.(15) reproduces the Ginzburg-Landau relations. In the quasi-classical regime of high enough temperatures and low magnetic fields, Eq.(15) reproduced both the descriptions of anisotropic 3D superconductivity and the Lawrence-Doniach model.

Our goal is to study theoretically the solutions of Eq.(15) under high magnetic fields that lead to the phenomenon of reentrant superconductivity [13-16]. Using the superconducting and band parameters for Li0.9MoO1.7, we can show that the reentrant phase can be achieved at fields of \( H \approx 100 \) Tesla at the transition temperature \( T^*(H = 100 \) T) \( \approx 1 \) K \( \approx T_c/2 \). To this end, we use the expansion of the Bessel function in Eq.(15) with \( 8t \epsilon/\omega_z \) as the small quantum parameter:

\[ J_0(\varepsilon) \approx 1 - \frac{4t^2 \epsilon^2}{\omega_z^2} \left[ 1 - \cos \left( \frac{\omega_z}{2v_F} (x - x') \right) \right], \quad (16) \]
where in the subsequent approximation the Bessel function expansion was averaged over \( x + x' \) variables. We use the defining relation for the zero-field critical temperature,

\[
\frac{1}{g} = \int_{d}^{\infty} \frac{2\pi T_c dz}{v_F \sinh \left( \frac{2\pi T_c}{v_F} z \right)},
\]

and the following standard approximation:

\[
\int_{0}^{\infty} \frac{1 \cdot \cos(\beta x)}{\sinh(x)} dx \approx \ln(2\beta \gamma),
\]

where \( \gamma \approx 1.781 \) is the exponential of the Euler constant. Substituting the relations in Eqs.(16)-(18) into the gap integral equation, and using the special trial solution \( \Delta(x) = \text{const.} \), we obtain an analytical equation that implicitly determined the critical reentrance temperature, \( T^*(H) \), as a function of magnetic field:

\[
\ln \left[ \frac{T_c}{T^*(H)} \right] = \frac{4t^2}{\omega_0^2(H)} \ln \left[ \frac{\gamma \omega_z(H)}{\pi T^*(H)} \right].
\]

For the solution (19) of the above equation, the numerical values \( t_z = 14K \) and \( \omega_z(H) = e v_F a_z H/c = 0.58H \) K/Tesla are obtained using the parameters for \( \text{Li}_{0.9}\text{Mo}_{6}\text{O}_{17} \) calculated in Ref. [21]. Thus, Eq.(19) can be numerically solved to obtain the field dependence of the reentrance transition temperature, shown in Fig.3. [Note that, in Fig.3, the superconducting transition temperature is shown only for high magnetic fields, \( H \geq 90 \) T, where the approximation (16) and, thus, Eq.(19) are valid. Nevertheless, at lower magnetic fields, superconductivity is still stable, although may have exponentially small transition temperature (see Refs.[13-16]).]

We recall that the superconducting transition temperature for \( \text{Li}_{0.9}\text{Mo}_{6}\text{O}_{17} \) in the absence of magnetic field is \( T_c = 2.2 \) K. Based on the above curve, we can see that the reentrant superconducting phase in \( \text{Li}_{0.9}\text{Mo}_{6}\text{O}_{17} \) can be obtained at the experimentally available nondestructive pulsed magnetic field and temperature of this configuration, we take the field and vector potential, along with the generalization of the Hamiltonian in Eq.(3) to be

\[
\vec{H} = (0, H \cos \alpha, H \sin \alpha), \quad \vec{A} = (0, H x \sin \alpha, -H x \cos \alpha),
\]

\[
\hat{H}^z = \mp i v_F \frac{d}{dx} - 2t_z \cos \left( p_z a_z + \frac{\omega_z(\alpha)}{v_F} x \right)
- 2t_y \cos \left( p_y a_y - \frac{\omega_y(\alpha)}{v_F} x \right). \quad (20)
\]
By following steps similar to the ones that lead to the derivation of Eq.(15), we can show that the generalization of the gap integral equation with a magnetic field inclined at angle \( \alpha \) takes the following form:

\[
\Delta(x) = \frac{g}{2} \int_{|x' - x| > d} \frac{2\pi T \Delta(x') dx'}{v_F \sinh \left[ \frac{2\pi T}{v_F} |x - x'| \right]} \times \nonumber
\]

\[
J_0 \left\{ \frac{8t_y}{\omega_y(\alpha)} \right\} \sin \left[ \frac{\omega_y(\alpha)}{2v_F} (x - x') \right] \sin \left[ \frac{\omega_y(\alpha)}{2v_F} (x + x') \right] \right\} \times 
\]

\[
J_0 \left\{ \frac{8t_z}{\omega_z(\alpha)} \right\} \sin \left[ \frac{\omega_z(\alpha)}{2v_F} (x - x') \right] \sin \left[ \frac{\omega_z(\alpha)}{2v_F} (x + x') \right] \right\}, 
\]

where the oscillation frequencies are now defined as

\[
\omega_y(\alpha) = \frac{ev_F a_y}{c} H \sin \alpha, \quad \omega_z(\alpha) = \frac{ev_F a_z}{c} H \cos \alpha. \quad (22)
\]

For this problem we consider Eq.(21) in the high-field reentrant regime, and introduce a normalized Ginzburg-Landau like trial solution with a variational parameter \( \kappa \):

\[
\Delta(x) = \left( \frac{2\kappa}{\pi} \right)^{1/4} e^{-\kappa x^2}. \quad (23)
\]

With a change of variables \( x' - x = (v_F / 2\pi T)z \) and the normalization condition \( \int \Delta^2(x) dx = 1 \), Eq.(21) can be recast in the form

\[
\frac{2}{g} = \int_{-\infty}^{\infty} dz \Delta(x + z) \Delta(x) \times \nonumber
\]

\[
J_0 \left\{ \frac{8t_y}{\omega_y(\alpha)} \right\} \sin \left[ \frac{\omega_y(\alpha)}{4\pi T} (2x + z) \right] \right\} \times 
\]

\[
J_0 \left\{ \frac{8t_z}{\omega_z(\alpha)} \right\} \sin \left[ \frac{\omega_z(\alpha)}{4\pi T} (2x + z) \right] \right\}, \quad (24)
\]

where \( d_1 = 2\pi T d / v_F \). At high magnetic fields and small angles, the first Bessel function in Eq.(24) can be expanded to leading term:

\[
J_0(\cdots) \approx 1 - \frac{t^2 \omega_y^2(\alpha)}{4\pi^2 T^4} \frac{\omega_z(\alpha)}{4\pi T} \approx x^2. \quad (25)
\]

The trial solution in Eq.(23) along with the expansion in Eq.(25) and Eq.(16) are substituted into Eq.(24) to evaluate in integrals and optimize the resulting expression with respect to \( \kappa \), giving the maximum transition temperature as a function of the angle: \( T^*(\alpha, H) \). Using the defining condition for zero-field critical temperature in Eq.(17), and the value of the integral

\[
\int_0^\infty \frac{z^2 dz}{\sinh(z)} = \frac{7\zeta(3)}{2},
\]

the resulting expression is

\[
\ln \left[ \frac{T^*(H)}{T^*(\alpha, H)} \right] = \left( \frac{v_F}{2\pi T^c} \right)^2 \frac{7\zeta(3)}{2} \left( \frac{\kappa}{2} + \frac{\lambda^2(\alpha)}{4\kappa} \right), \quad (26)
\]

where \( T^*(H) = T^*(\alpha = 0, H) \), and

\[
\lambda^2(\alpha) = \frac{4t^2 \omega_y^2(\alpha)}{v_F^2}. \quad (27)
\]

with \( \omega_y(\alpha) \) given in Eq.(22), and \( \zeta(3) \approx 1.202 \) being the value of the Riemann zeta function. The value of \( \kappa \) that minimizes the expression in Eq.(26), thus giving the maximum transition temperature, \( T^*(\alpha, H) \), is

\[
\kappa = \frac{\lambda(\alpha)}{\sqrt{2}}. \quad (28)
\]

For small angles, \( \alpha \), we use the approximation

\[
\ln \left[ \frac{T^*(H)}{T^*(\alpha, H)} \right] \approx \frac{T^*(H) - T^*(\alpha, H)}{T^*(H)}, \quad (29)
\]

and substitute the result of Eq.(28) into Eq.(26) to solve for \( \omega_y(\alpha) \) and obtain an expression for the following dependence of \( T^*(\alpha, H) \), in the high magnetic field regime corresponding to the reentrant superconducting phase:

\[
T^*(\alpha, H) = T^*(H) \left( 1 - \frac{H}{H_{GL}} \sin \alpha \right), \quad (30)
\]

where

\[
H_{GL} = \frac{4\sqrt{2\pi^2 c T^2}}{7\zeta(3) t y v_F a_y} \quad (31)
\]

is the Ginzburg-Landau upper critical field for \( \alpha = 0^\circ \). [Note that the obtained angular dependence (30),(31) has a different meaning than the standard Ginzburg-Landau one since in our case it is valid only at high magnetic fields.]

The results of Eqs.(30),(31), along with the value of \( T^*(\alpha = 0) \approx 1 K \) at \( H \approx 100 T \) can be used to calculate the angular dependence of the reentrant transition temperature, plotted in Fig.5. We note that the sharp drop observed in the transition temperature for angles near \( \alpha = 0 \) shows that one needs a careful alignment of a magnetic field during the corresponding experiment. As follows from Fig.5, the accuracy of the alignment of the field has to be better than \( \delta \alpha = 0.20^\circ \) in \((y, z)\) plane (see Fig.4). On the other hand, it is known that magnetic fields of order of \( H = 15 T \) can destroy superconductivity in \( Li_{0.3}Mo_{0.17} \) when the field is aligned parallel to the conducting chains (see Ref. [19]). Therefore, parallel to the chains component of the magnetic field has
FIG. 5: Angular variation of reentrant superconducting transition temperature (in Kelvin), $T^*(\alpha)$, at $H = 100 \, T$ for small inclination angles, $\alpha$ (in degrees) from the optimal experimental geometry.

to be less than $\delta H_\parallel \simeq 5 \, T$ in the experiments. Thus, accurate angular orientation is important for detection of the reentrant superconducting phase at fields of order of $H \simeq 100 \, T$, where small inclination of the field (in particular, towards the $z$ axis) can destroy superconductivity.

IV. CONCLUSION

In this paper, we have studied the quantum limit reentrant superconductivity phenomenon in the layered Q1D conductor Li$_{0.9}$Mo$_6$O$_{17}$. Our results show that superconductivity can be restored and potentially experimentally detected in this compound at the reentrant transition temperature $T^*(H = 100 \, T) \simeq 1 \, K$ when a field of order $H \simeq 100 \, Tesla$ is aligned parallel to the layers and perpendicular to the conducting chains. We noted that such magnetic fields are currently experimentally available as pulsed non-destructive fields. Furthermore, we have specified how the reentrance transition temperature, $T^*(\alpha, H)$ varies for arbitrary, as well as small angular deviations from the optimal experimental geometry. This information is important for accurate alignment of a sample in magnetic field. If confirmed experimentally, the reentrant superconducting phase in Li$_{0.9}$Mo$_6$O$_{17}$ would be the first example of survival of superconductivity in ultra-high magnetic fields and would in addition unequivocally confirm spin-triplet pairing nature in this compound.

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