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## Back to the problem of the upper critical fields in organic superconductors

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**Résumé.** — Nous montrons que les valeurs des champs critiques supérieurs des supraconducteurs organiques peuvent être interprétées dans le cadre de la théorie des supraconducteurs propres de type II. L'observation de valeurs dépassant la limite de Clogston pourrait indiquer l'existence d'une supraconduction avec couplage triplet.

**Abstract.** — It is shown that the properties of the upper critical fields in organic superconductors can be understood in terms of pure type II superconductivity. The experimental observation of high critical fields violating Clogston's criterion could give support in favour of triplet-paired superconductivity.

In this short Letter we shall reinvestigate the theoretical interpretation of the experimental observed upper critical fields,  $H_{c2}$ , in the Bechgaard salt superconductors  $(\text{TMTSF})_2\text{X}$ . We want to comment as to whether evidence in favour of the Pauli limitations (the Clogston criterion) are really present in these materials. Owing to the rather large values of the critical fields  $H_{c2}$  obtained in these superconductors [1], the data has been interpreted in terms of the so-called dirty limit of the BCS-like superconductors [2]. Meanwhile, there is strong evidence that these materials can be extremely clean, which is inconsistent with this approach. Thus, the concentration of defects, as estimated from the irradiation effect on the transition temperature  $T_c$  (and on the residual resistance) gives approximately 100 ppm of defects per molecule [3]. In other words, the mean free path along the chain (i.e. the  $a$ -direction) is  $l > 10^4 a$ , where  $a$  is the lattice constant. Taking the tight binding estimate for the electron velocity,  $v_a$  :

$$v_a = \frac{at_a}{\hbar\sqrt{2}} \quad (1)$$

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and  $t_a \approx 2000$  K [4], one immediately obtains

$$\hbar/\tau \lesssim 0.1 \text{ K}$$

while  $T_c$  is about 1 K.

Because of these reasons we adopt the point of view that these superconductors are of the IIInd type already in the pure limit, and we shall try to use the microscopic expressions for the effective mass tensor in the Ginzburg-Landau equation (near  $T_c$ ) derived in reference [5].

$$\frac{1}{m_{ik}} = \frac{7 \zeta(3)}{12 \pi^2 T} \langle v_i v_k | \psi^2 | \rangle. \quad (2)$$

(Here the average is taken over the Fermi surface,  $\psi(k)$  is the normalized superconductivity order parameter,  $\langle | \psi^2 | \rangle = 1$ .)

Using the orthorhombic approximation for the (TMTSF)<sub>2</sub>X-lattice and the tight binding dispersion law for all three directions (which is useful for the purpose of semiquantitative estimations),

$$\varepsilon(\vec{p}) = 2 t_a \cos \tilde{p} \frac{\tilde{a}}{2} + 2 t_b \cos \tilde{p} \tilde{b} + 2 t_c \cos \tilde{p} \tilde{c}, \quad (3)$$

one gets the following « mass-tensor » :

$$\begin{aligned} \frac{1}{m_a} &= \frac{7 \zeta(3) t_a^2 a^2}{24 \pi^2 T_c \hbar^2} \\ \frac{1}{m_b} &= \frac{7 \zeta(3) t_b^2 b^2}{6 \pi^2 T_c \hbar^2} \\ \frac{1}{m_c} &= \frac{7 \zeta(3) t_c^2 c^2}{6 \pi^2 T_c \hbar^2}. \end{aligned} \quad (4)$$

A variation of the factor  $| \psi |^2$  along the F.S. in equation (2) has been omitted in the evaluation of equation (4). Since  $| \psi |^2$  varies along the F.S., including its variation would increase the resulting fields,  $H_{c_2}$ .

As the Cooper pair charge equals  $2e$ , one would immediately get the well known result of the G.-L. theory :

$$\frac{e \hbar H_{c_2}^i}{c^* \sqrt{m_k m_j}} = T_c - T \quad (5)$$

where  $c^*$  in (5) is the velocity of light, and  $i \neq k \neq j$  correspond to the three mutually orthogonal directions. Substitution of (4) into (5) leads to the results :

$$\begin{aligned} H_{c_2}^a &= \frac{T_c - T}{T_c} \frac{6 \pi^2 c^* \hbar T_c^2}{7 \zeta(3) e t_b t_c b c} \\ H_{c_2}^b &= \frac{T_c - T}{T_c} \frac{12 \pi^2 c^* \hbar T_c^2}{7 \zeta(3) e t_a t_c a c} \\ H_{c_2}^c &= \frac{T_c - T}{T_c} \frac{12 \pi^2 c^* \hbar T_c^2}{7 \zeta(3) e t_a t_b a b}. \end{aligned} \quad (6a)$$

Equations (6a) can also be rewritten in a more practical form

$$\begin{aligned} H_{c_2}^a &= \frac{4.84 \times 10^4}{t_b t_c} T_c (T_c - T) \\ H_{c_2}^b &= \frac{9.68 \times 10^4}{t_a t_c} T_c (T_c - T) \\ H_{c_2}^c &= \frac{19.3 \times 10^4}{t_a t_b} T_c (T_c - T) \end{aligned} \quad (6b)$$

where the magnetic fields are given in kOe and  $t_i$  in kelvin.

Taking the transverse critical field data of  $(\text{TMTSF})_2\text{ClO}_4$  (R-state,  $T_c = 1.2$  K) reported by Ishiguro *et al.* [6], namely  $dH_{c_2}^b/dT = 12.5$  kOe/K and  $dH_{c_2}^c/dT = 0.8$  kOe/K, and using the following set of band parameters  $t_a = 2000$  K,  $t_b = 200$  K and  $t_c = 5$  K [7], one would derive from equations (6b)  $dH_{c_2}^b/dT = 11.6$  kOe/K and  $dH_{c_2}^c/dT = 0.6$  kOe/K. The latter values lie, admittedly, within the accuracy of the central field experimental determination (considering the approximation in Eq. (3)).

With these parameters in mind we now turn to the values of  $H_{c_2}^a$  which are possible at low temperatures in the pure limit. This would give  $dH_{c_2}^a/dT = 58$  kOe/K, and probably even more if  $|\psi|^2$  in equation (2) is taken into account.

Such large values of  $H_{c_2}^a$  are consistent with the little existing experimental data for  $(\text{TMTSF})_2\text{ClO}_4$ ; for example, an early experiment with  $T_c = 0.86$  K [3] (the sample was not properly in a R-state) has led to  $dH_{c_2}^a/dT \approx 50$  kOe/K while an other study with  $T_c = 1$  K [8] has provided  $dH_{c_2}^a/dT \approx 38$  kOe/K. Measurements of transverse fields have also been reported in  $(\text{TMTSF})_2\text{AsF}_6$  under pressure of 11 kbar [1] with slopes  $dH_{c_2}^a/dT$  of 16.6 kOe/K and 1 kOe/K for  $b$  and  $c$ -directions respectively.

At this point we would like to emphasize that the precise determination of the critical fields along the  $a$ -direction meets several complications, especially within a high pressure vessel at very low temperature. The correct alignment of the sample along the magnetic field is only one of the problems. But there are also problems regarding the low symmetry of the  $(\text{TMTSF})_2\text{X}$  phases (the angle between  $a$  and  $a^*$  axis is about  $190^\circ$ ) and the possibility of twinning among  $(\text{TMTSF})_2\text{X}$  crystals.

The quality of data for  $(\text{TMTSF})_2\text{PF}_6$  under pressure is too low to allow any definite conclusions. The  $T_c$  of 0.8 K under 11 kbar [3] is different from other reported results existing in the literature [2].

The values of  $H_{c_2}^a$  mentioned above exceed the limitation imposed by the Pauli pair-breaking mechanism by a factor about two ( $2 \mu_B H_{\text{Pauli}}(0) = 3.55 T_c$ ). The spin-orbit interactions violating the applicability of the Clogston criterion in its simplest form are probably not important due to a relatively weak spin-orbit coupling and the minor presence of impurities. In this respect, superconductivity compounds built around sulfur-based organic molecules seem to show similar behaviours [9], but the quality of samples is not as good as in the Bechgaard series.

Thus, we conclude that the orbital mechanism destroying superconductivity in the presence of a large magnetic field is able to provide the correct order of magnitude for  $H_{c_2}$  along the  $a$ -direction, even in the pure limit, and in addition, the large values of fields are not suppressed by the Pauli mechanism. We claim that the critical fields in  $(\text{TMTSF})_2\text{X}$  superconductors can be explained in the present pure-limit picture, as well as in the dirty-limit which was adopted for previous interpretations. This last fact could be considered as an indication in favour of « triplet superconductivity ». Furthermore, as indicated by  $H_{c_2}$  measurements close to the SDW state in  $(\text{TMTSF})_2\text{AsF}_6$  at 9.5 kbar [1], critical fields seem to be somewhat enhanced and  $T_c$  depressed by the proximity to the SDW state.

It seems worthwhile emphasizing once again that the previous arguments were based on the Fermi surface picture using a single-particle approach, say, to the conductivity. Meanwhile, there are indications that this may not be valid [2]. As an example, we mention the very unusual behaviour of the magnetoresistance at low temperature (at  $T \approx 4$  K) and at low fields ( $H \lesssim 10$  kG) which is very large,  $\Delta\rho/\rho \sim 1$ , and is strongly dependent on the field orientation.

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