Superconductivity in the charge-density-wave state of the organic metal
\( \alpha-(\text{BEDT-TTF})_2\text{KHg(SCN)}_4 \)

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The superconducting transition in the layered organic compound \( \alpha-(\text{BEDT-TTF})_2\text{KHg(SCN)}_4 \) has been studied in the two hydrostatic pressure regimes where a charge-density wave is either present or completely suppressed. Within the charge-density-wave state the experimental results reveal a network of weakly coupled superconducting regions. This is especially seen in a strong enhancement of the measured critical field and the corresponding positive curvature of its temperature dependence. Further, it is shown that on lowering the pressure into the density-wave state traces of a superconducting phase already start to appear at a much higher temperature.

INTRODUCTION

The organic metal \( \alpha-(\text{BEDT-TTF})_2\text{KHg(SCN)}_4 \) has already raised great attention due to a variety of novel physical phenomena found in its low-temperature charge-density-wave (CDW) state\(^1\)\(^2\)\(^3\)\(^4\). Of particular interest have been, for example, new kinds of modulated CDW states existing in magnetic fields above the paramagnetic limit\(^5\)\(^6\)\(^7\)\(^8\) and phase transitions induced by high magnetic fields due to a specific interplay between the Pauli paramagnetic and orbital effects\(^5\)\(^9\). Apart from the high-field phenomena there are other interesting properties, such as the coexistence/competition of CDW and superconductivity which have not been thoroughly addressed so far.

Owing to a strongly anisotropic electron system, the Fermi surface (FS) of this compound consists of coexisting open sheets and cylinders\(^10\)\(^11\). The slightly warped sheets correspond to a quasi-one-dimensional (Q1D) electron band. The latter emerges due to an enhanced electron transfer integral \( t_a \) in the crystallographic \( a \)-direction between the organic BEDT-TTF molecules resulting in a chain-like coupling within the conducting \( a-c \) plane\(^11\). At about 8 K there is a phase transition to the CDW state\(^11\)\(^2\)\(^2\)\(^2\)\(^2\)\(^2\)\(^2\), in which these sheets of the FS become nested and the Q1D carriers are gapped. The system, however, keeps its metallic character due to the second, quasi-two dimensional (Q2D) band.

Remarkably, the iso-structural salt \( \alpha-(\text{BEDT-TTF})_2\text{NH}_4\text{Hg(SCN)}_4 \) (hereafter we refer to both compounds as NH\(_4\)- and K-salt) does not undergo the density wave transition but instead becomes superconducting (SC) at \( \approx 1 \) K\(^13\)\(^14\). The absence of a density wave is interpreted to be due to a higher inter- to intrachain-coupling ratio \( t_c/t_a \) of the organic molecules within the layers, that strongly deteriorates the nesting conditions of the open sheets of the FS\(^16\)\(^17\). Moreover, it has been shown\(^17\) that by tuning the ratio of the lattice constants \( c/a \) under uniaxial strain a density wave can be even (i) induced in the NH\(_4\)-salt and (ii) suppressed in the K-salt, a SC state being stabilized at \( \approx 1 \) K. Based on combined uniaxial strain measurements and band structure calculations Kondo et al.\(^16\) have proposed that the major contribution to superconductivity comes from the Q1D band.

Similarly, hydrostatic pressure turns out to worsen the nesting conditions in the K-salt\(^16\). The increase of the interchain coupling leads to a decrease of the density wave transition temperature, and at the pressure \( P_c \approx 2.5 \) kbar the density wave is completely suppressed, a normal metallic (NM) state being stabilized\(^9\). Hydrostatic pressure studies\(^15\) have also revealed superconductivity in the K-salt but at temperatures much lower than it was observed in the uniaxial strain experiments. Remarkably, the superconductivity was shown to persist over the whole pressure range studied, from 0 up to 4 kbar, i.e. it exists both in the NM and in the CDW regimes. This offers a direct opportunity to study the influence of a CDW on a SC system.

Basically, the SC pairing competes with the density-wave instability for the FS\(^14\)\(^22\). Therefore one would expect the SC transition to be suppressed upon entering the CDW region of the phase diagram since the Q1D carriers, which are supposed to be responsible for superconductivity\(^16\), become completely gapped below \( P_c \). On the other hand, it was predicted recently\(^21\)\(^22\) that density-wave fluctuations can even stimulate the SC pairing in the vicinity of the CDW ground state.

In this paper we present experimental studies of the SC transition in the K-salt at different pressures, temperatures and magnetic fields. We argue that below the critical pressure \( P_c \) the SC phase exists in the form of an array of weakly coupled small SC regions or filaments embedded in the metallic CDW matrix. Moreover, we show that the SC onset temperature becomes drastically enhanced on lowering the pressure across the CDW/NM boundary which is likely a sign of a nontrivial effect of the CDW on the superconductivity in this compound.
EXPERIMENT

The main results presented in the paper were obtained from interlayer resistance measurements using the standard four probe geometry and a.c. measuring technique. Two samples, hereafter referred to as samples #1 and #2, were measured simultaneously. The samples had the dimensions of $0.6 \times 0.5 \times 0.2 \text{ mm}^3$ and $1.0 \times 0.3 \times 0.05 \text{ mm}^3$, respectively, the smallest dimension being in the interlayer direction. Additionally, measurements with the current applied along the biggest dimension, i.e. nominally parallel to the layers, were done on sample #2. Of course, due to the extremely high anisotropy of our compound this measured “inplane” resistance includes a mixture of the intra- and interlayer components of the resistivity tensor [23]. To minimize the influence of the interlayer component the thinnest sample was chosen. The in- and interplane resistances were measured in the same run by using the standard 6-probe geometry (four contacts were made to one of the biggest surfaces of the plate like sample and two contacts to the opposite surface). Thus, after comparing the measured in- and interplane resistances we were able to make reasonable conclusions about the temperature dependence of the intralayer resistivity.

Hydrostatic pressure was applied using a conventional berillium-copper clamp cell. The latter was mounted on a dilution refrigerator allowing the sample to be cooled down to 20 mK. The pressure value at low temperatures was determined from the resistance of a calibrated manganin coil to an accuracy better than $\pm 100 \text{ bar}$.

At the lowest temperatures, special care was taken to control and minimize overheating due to the transport current and field-sweep induced eddy currents. On measuring the interlayer resistance with the applied current of 50 nA the overheating of the sample was found to be $< 5 \text{ mK}$ at 20 mK. The sweep rates of the magnetic field were chosen extremely low, $\approx 1 \text{ mT/min}$, so that eddy currents had no visible effect on the sample temperature.

Further, since the SC properties are extremely sensitive to magnetic fields, the superconducting magnet used in the experiment was always carefully demagnetized before the measurements, so that the remanent field was below 0.5 mT.

RESULTS AND DISCUSSION

Resistive SC transition at zero field

In Fig. 1 several temperature sweeps of the interlayer resistance for sample #1 measured at different pressures show the already reported behavior [18]. At $P = 3 \text{ kbar}$ the resistance exhibits a normal metallic behavior on cooling until at 110 mK a sharp SC transition ($\Delta T \approx 10 \text{ mK}$) occurs. Above $P_c \approx 2.5 \text{ kbar}$ the SC transition remains sharp and the critical temperature $T_c$, defined as the midpoint of the transition, shows a negative pressure dependence of about $-30 \text{ mK/kbar}$ [13]. This value is 1-2 orders of magnitude lower than measured in other BEDT-TTF-based superconductors, where a strong linear suppression of superconductivity with hydrostatic pressure is commonly observed [14, 24].

Kondo et al. [16] performed uni-axial strain experiments on the NH$_4$-salt, with a combined X-ray determination of the lattice parameters. Their tight binding band structure calculation proposed the changes of the SC transition temperature to be reasonably described by the changing density of states (DOS) at the Fermi level within the BCS model. However, they mention that such a simple description fails as one approaches the density wave state. Under hydrostatic pressure, the pressure dependence of $T_c$ in the K-compound is found to be an order of magnitude lower than observed [27] in the NH$_4$-salt. This is quite unusual: normally isostructural organic superconductors with different anion layers display approximately the same pressure dependence of $T_c$ [19].

Thus, also in the hydrostatic pressure case the proximity to the density-wave instability in the K-salt seems to affect the SC transition in the metallic state. A direct comparison of the SC properties between the two compounds may, therefore, be inappropriate. Indeed, the value $dT_c/dP = -30 \text{ mK/kbar}$ is closer to that observed in the Q1D TMTSF (or TMTTF) based organic metals in which the SC state exists in the hydrostatic pressure range right next to the spin-density-wave state [19].

Obviously, in the vicinity of a density-wave transition a detailed consideration of different carrier interactions,
due to which different instabilities of the metallic ground state compete with each other, becomes necessary.

As mentioned in the introduction, on entering the CDW state, i.e. with lowering the pressure below $P_c$, the superconductivity does not vanish. At 2.5 kbar $T_c$ remains at the value observed at 3 kbar, instead of further increasing, as would be expected from an extrapolation from higher $P$. With further decreasing the pressure, the transition broadens and gets a kind of a step-like structure as can be seen in Fig. 1. This leads to a strong suppression of the temperature $T_0$ at which zero resistance is reached; at ambient pressure the resistance does not vanish down to 20 mK. Thus, there is a clear effect of the CDW on the resistive SC transition. We note that the observed data are also very well in line with the former proposal \[ P_c = 2.5 \text{ kbar} \] for the complete suppression of the CDW state.

The overall behavior described above was also observed on sample #2, measured simultaneously. The superconducting transition temperature, however, appears to be sample dependent. The difference between the resistively measured transition temperatures of samples #1 and #2 is approximately 10% at $P > 2.5$ kbar, and becomes even stronger in the CDW state, at $P \leq 2.5$ kbar. This suggests the impact of the CDW on the superconductivity to be also dependent on impurities or defects.

In Fig. 2 a comparison between the in- and interplane resistances is shown for sample #2 at pressures above and below $P_c$. Note that in order to measure the inplane resistance to a reasonable accuracy the applied current had to be at least 0.5 $\mu$A. However, despite this high current, that caused a small, $\sim 1$-2 mK, overheating at the transition temperature, it is seen that the SC transition in the plane occurs at a higher temperature in comparison to the interlayer one. This difference in the transition temperatures originates most likely from the layered character of superconductivity: the SC ordering is first established within the layers whereas the interlayer coherence develops at lower temperatures. Such a scenario has also been proposed for the NH$_4$-compound \[ P_c = 2.5 \text{ kbar} \] where the interlayer coherence length $\xi_{\perp}$ is found to be smaller than the interlayer spacing of 20 Å \[ P_c > 2.5 \text{ kbar} \]. This can also be assumed for the K-salt, since, although $T_c$ is here an order of magnitude lower, the in- to interplane anisotropy of the Fermi velocity is considerably higher than the one in the NH$_4$ compound \[ 2.5 \text{ kbar} \].

At 1.85 kbar the inplane resistance is zero below 50-60 mK whereas the interlayer transition does not vanish down to the lowest temperature. A clear broadening of the inplane transition within the CDW state is, however, also observed. We therefore presume that the evolution, with pressure, of the SC transition in the intralayer resistance is similar to that described above for the interlayer resistance. This is supported by the previous report by Ito et al. \[ 2.5 \text{ kbar} \] on the incomplete transition in the inplane resistance at ambient pressure.
We now discuss a possible reason for broadening the SC transition. First, we note that for all measured samples the transition width is maximum at zero pressure and decreases as the pressure is increased until the critical value $P_c$ is reached; at $P > P_c$ the transition width is relatively small and approximately constant, $\Delta T_c \approx 10$ mK. Thus, the broadening cannot be ascribed to pressure inhomogeneity. Generally one can think of phase fluctuations, typical of highly anisotropic electron systems with small superfluid density, that leads to a suppression of the bulk superconductivity [28] as has been observed in high $T_c$ superconductors [20]. However, in our system the SC transition temperature is of the order of 100 mK. In this case, the zero-temperature phase-stiffness of superconductivity is high enough, so that effects of phase fluctuations on $T_c$ are negligible [29].

A clue to finding the real nature of the strongly broadened resistive transition lies in a comparison of transport and magnetization measurements. In Fig. 3 we show the temperature dependence of the interlayer resistance for three different samples at ambient pressure. As can be seen, sample #3 almost reaches zero resistance on cooling down to 20 mK, reflecting the already mentioned sample dependence of the SC transition [18]. This, however, does not mean that the whole sample at lower temperatures is in the SC state. D.C. magnetization measurements on the same sample made on a SQUID magnetometer could not resolve any Meissner effect, even down to 6 mK. Therefore, the zero resistance most likely originates from a network of weakly coupled SC regions or filaments. Thus, we suggest that the SC and CDW phases are separated in space. This is also supported by theoretical predictions that a CDW leads to a suppression of superconductivity [20]. We consider an inhomogeneous system of SC islands embedded in a metallic (actually CDW) matrix to be more likely. The SC coherence, thus, develops within the islands until at lower temperatures they couple to each other via the proximity effect, providing a percolation network. At ambient pressure the islands are strongly separated, so that a completely coupled system does not exist at $T > 20$ mK. A strong broadening of the “bulk” SC transition is indeed known to exist in a two dimensional array of SC islands which are embedded in a metallic matrix [30, 31]. After the islands become SC the decrease of the resistance is determined by the growth of the normal metallic coherence length on lowering the temperature, i.e. the proximity effect. Since we have no possibility at the moment to study the magnetization under pressure, we cannot directly verify the absence of the Meissner effect. However, as we shall see next, the inhomogeneous nature of superconductivity under hydrostatic pressure is supported by measurements of the SC transition in magnetic fields.

Magnetic field effect

In Fig. 4 we show the magnetic field sweeps made on sample #1, with the field directed perpendicular to the planes, at different temperatures and two pressures, above and below the critical value $P_c$. At zero field the transition temperature at these two pressures is approxi-
also occur in our compound. This means that there’s a

ture of the upper critical field. A similar scenario might

with lowering than the coherence length \[29\]. A dimensional crossover

sions perpendicular to the field direction becomes less

enhanced in a superconductor if at least one of the dimen-

\[\approx\]

where the resistance is

the inflection point (circles), and the end of the SC transiti-

\[P\]

\[=3.5\text{ kbar (open symbols) determined from the resistive}

transition in the field sweeps using different criteria. The

\[P\]

\[=2\text{ kbar (solid symbols) and}

\[P\]

\[=3.5\text{ kbar (open symbols) determined from the resistive}

transition does not necessarily match the upper critical field

\[H\]

\[c_2\] at \(P < P_c\). As was argued above, the superconduc-

tivity is most likely inhomogeneous in this pressure range.

Therefore, the resistive transition may be largely deter-

mined by the coupling between the randomly separated

SC islands rather than by \(H_{c2}\) inside the islands. This

means that not only the value of \(H_p\) defined above can

differ from the real \(H_{c2}\) but also its temperature depen-

dence. Although an exact theoretical description of the

resistive transition of a proximity coupled random array

of SC islands in a magnetic field still has to be worked

out, a comparison to existing inhomogeneous supercon-

ductors shows that a strong positive curvature of \(H_p\) can

be expected.

As an example one can mention polymeric sulfur nit-

tride (SN), a compound that consists of bundles of SC

filaments. For a magnetic field applied perpendicular to

the fiber axis the temperature dependence of the resis-

tive transition was shown to exhibit a positive curvature

\[82\]. Another, and probably more relevant example is

the well known CDW compound NbSe_3. It has been re-
-reported \[33\] that within the CDW state of NbSe_3 a small

fraction of the sample becomes SC and it has been pro-

posed to emerge within the boundaries of CDW domain

walls, where the CDW order parameter is supposed to

become zero. This would then indeed be a system of SC

regions separated by the metallic CDW phase similarly to

our present case. At higher pressures the CDW gap

becomes smaller and the domain wall fraction, where un-
gapped Q1D electrons exist, is expected to become big-

ger. Moreover, a strong sample dependence of the SC

properties would not be surprising in such a model, since

crystal defects or impurities very likely affect the domain

structure. Whether such a domain structure really exists

in the title compound we cannot judge from our data,

but the similarities between both compounds with re-

spect to their SC properties suggest the nature of the

critical field behavior to be the same. The possibility

of domains within a Q1D CDW system has indeed been

predicted \[34\]. Furthermore, Gor’kov et al. mention that

the superconductivity would be expected to survive in

the domain walls perpendicular to the conducting chain

direction \[34\].

Noteworthy, there might exist a narrow pressure region

in the vicinity of \(P_c\), in which the system becomes inho-

mogeneous, irrespective of the CDW domain structure

\[36\]. Such an inhomogeneous system, associated with a

first order phase transition, was also shown to have an

enhanced SC upper critical field \[37\] in the spin density

wave compound (TMTSF)_2PF_6.
temperatures than in the NM state (decrease of the interlayer resistance accelerates at much higher temperatures at pressures \( \leq 2 \) kbar). This decrease strongly depends on the level of the applied current and field. With increasing the current or field the resistance decrease becomes suppressed. Note that the main transition shifts only slightly at higher currents in Fig. 8. Therefore, effects of overheating can be neglected.

The present data manifest that traces of superconductivity, occupying a small fraction of the crystal volume, exist already at much higher temperatures. The described behavior was found throughout the entire CDW pressure range. The onset temperature of superconductivity is \( \approx 0.22 \) K at 2.5 kbar and 0.30 K at 2 kbar and 0 kbar. These findings were reproduced on several samples. They are also consistent with the ambient pressure results of Ito et al. [27].

By contrast to the CDW pressure region, in the NM state such an accelerated decrease of the resistance above the bulk SC transition has not been detected (see the 3 kbar curve in Fig. 8). Hence, we conclude that the dramatic increase of the SC onset temperature is a consequence of entering the CDW region of the phase diagram. The whole \( P-T \) phase diagram including all phases must, therefore, look as depicted in Fig. 9. Since the SC transition is sharp above \( P_c \) but becomes broadened in the CDW region, we take here the midpoint for the NM/SC transition (filled triangles in Fig. 9) and the onset and zero-resistance temperatures for the main SC transition in the CDW state (open triangles and circles, respectively). The onset temperature of small SC regions in the CDW state (filled squares) is determined by the inflection point in the temperature dependent resistance. Obviously, there is an extended range in the \( P-T \) phase diagram. Filled symbols show the phase transitions between different states. Open symbols mark the onset and zero-resistance temperatures of the broadened main SC transition in the CDW state. The lines are guides for the eye.

**Enhanced SC onset temperature**

Besides the broadening of the main SC transition, all temperature sweeps at pressures \( \leq 2.5 \) kbar show an unusually strong decrease (negative curvature) of the resistance in a remarkably wide temperature range well above the \( T_c \) value that would be expected from its linear extrapolation from \( P > P_c \). Fig. 8 shows, in an enlarged scale, the resistance of sample \#1 at 2 kbar, at temperatures right above the main transition, which is still rather sharp at this pressure. For comparison, the 3 kbar resistance is also shown in the upper panel. As can be seen from the figure, the decrease of the resistance strongly depends on the level of the applied current and field. With increasing the current or field the resistance decrease becomes suppressed. Note that the main transition shifts only slightly at higher currents in Fig. 8. Therefore, effects of overheating can be neglected.

![FIG. 7: Temperature dependent interlayer resistance at different constant magnetic fields, at \( P = 2 \) kbar.](image7.png)

![FIG. 8: Fig. 8. Within the CDW state \( (P = 2 \) kbar) the decrease of the interlayer resistance accelerates at much higher temperatures than in the NM state \( (P = 3 \) kbar). This decrease strongly depends on the level of the applied current \( (a) \) and magnetic field perpendicular to the layers \( (b) \).](image8.png)

![FIG. 9: Proposed \( P-T \) phase diagram. Filled symbols show the phase transitions between different states. Open symbols mark the onset and zero-resistance temperatures of the broadened main SC transition in the CDW state. The lines are guides for the eye.](image9.png)
diagram that includes both ground states, superconductivity and density wave.

If the superconductivity is indeed spatially restricted to the CDW domain boundaries, as suggested above, one can understand, why the CDW does not completely suppress the SC state, in contradiction to what has been theoretically proposed \[24\]. This will, however, not explain the enhanced SC onset temperature. In principle, in the model above one would still expect the opposite effect, namely that the SC island has a reduced onset temperature due to the proximity effect. On the other hand, we do not know in what way the superconductivity, located in the domain boundaries where the order parameter of the density wave reaches zero, is influenced by the CDW neighborhood. An interesting scenario to consider would be an additional stimulation of superconductivity in the CDW domain walls, such as, for example, a charge-fluctuation mediated pairing \[21\] \[22\] \[38\]. More investigations on this topic are highly desirable.

**CONCLUSION**

In conclusion, pronounced differences in the superconducting properties are observed between the CDW and the NM pressure regions. The determined phase diagram further confirms that \( P_c \approx 2.5 \) kbar is the critical pressure at which the CDW state becomes completely suppressed. Below \( P_c \), the broadening of the resistive SC transitions, the absence of the Meissner effect as well as the pronounced enhancement and positive curvature of the critical magnetic field point to the formation of a network of coupled SC regions embedded in the CDW matrix. We propose that the superconductivity is located within CDW domain walls. Furthermore, it is found that traces of a SC phase exist in the CDW region already at temperatures much higher than expected from the NM state. The origin of this remarkable and unexpected expansion of the SC temperature range remains at present one of the most intriguing questions.

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