J. Phys. IV ***France*** **12** (2002)
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DOI: 10.1051/jp4:20020394

Coexistence of superconductivity and spin density wave orderings in Bechgaard and Fabre salts

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Abstract. In a small range of pressure, superconductivity (SC) and Spin Density Wave (SDW) states are shown to coexist in the Bechgaard salt $(TMTSF)_2PF_6$ and the Fabre salt $(TMTTF)_2BF_4$. In $(TMTSF)_2PF_6$, a precise investigation of the (P,T) phase diagram has led us to demonstrate the coexistence of the two phases with a superconducting critical temperature which is pressure independent while the critical current at zero field is strongly depressed as the pressure is decreased. In $(TMTTF)_2BF_4$, using non-linear transport measurements, we present the signature of the presence of 1D superconducting filaments in a small range of pressure. We also investigate the compound under a magnetic field applied along the c*-axis : the upper critical field is more or **less** pressure independent and is about 2 Tesla (at zero temperature). We suggest that such a high critical field is compatible with the penetration of the magnetic field in the insulating regions of the compound in a similar way of Josephson vortices in layered superconductors.

1. INTRODUCTION

Quasi-one-dimensional organic compounds of the $(TM)_2X$ family where TM is either the tetramethyl-tetrathiofulvalene TMTTF or **tetramethyl-tetraselenafulvalene** TMTSF organic cations and X is an inorganic monoanion, can be described by a unified (Pressure, Temperature) phase diagram considering a different origin on the pressure scale for each salt. A one dimensional regime (1D) prevails in the upper left part (high temperature and low pressure region of the sulfur compounds) characterized by Mott-Hubbard localization, while spin density wave (SDW) and superconductivity (SC) compete at low temperatures and high pressures[1, 2]. This question **of** competition of a magnetic state and a superconducting (or even only metallic) state is actually intensively studied in cuprates [3] and has been also recently adressed in the κ-$(BEDT\text{-}TTF)_2X$ family.[4]

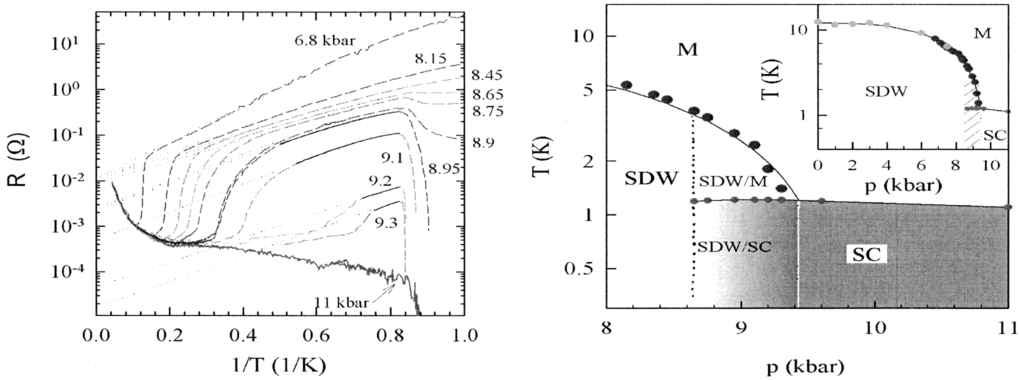
We first report experimentally SDW-SC phase segregation in a narrow range of pressure in the $(TMTSF)_2PF_6$ salt. Such a behavior is also observed in $(TMTTF)_2BF_4$ which is shown to present superconductivity at least in the pressure range 33.5-37.5 kbar. In both compounds, the inhomogeneous coexistence has been established from critical current measurements. In the $(TMTTF)_2BF_4$ salt, the upper critical field is shown to be large, H_{c2}~2Tesla, which certainly indicates the penetration of the field in the insulating regions in a similar way to Josephson vortices in lamellar superconductors.

2. EXPERIMENTAL

Low hydrostatic pressure was applied on $(TMTSF)_2PF_6$ in a regular beryllium-copper cell with silicon oil inside a Teflon cup as the pressure transmitting medium. The pressure was measured using an InSb pressure gauge located inside the cell close to the sample. High hydrostatic pressure, applied on $(TMTTF)_2BF_4$, was provided in a clamped cell made of non magnetic NiCrAl alloy with petroleum in a teflon cell as the pressure transmitting medium. In order to control the pressure when it is varied (only at room temperature), we have used a manganin pressure gauge located in the cell close to the sample. The pressure variation was controlled with an accuracy of around 100 bar. We have neglected the pressure loss during cooling assuming that it should be small and identical for all studied pressures. The pressure cells could be cooled down to 0.35 K (He^3 refrigerator) and eventually placed in a magnetic field up to 12 T. Longitudinal resistivity measurements were performed on single crystals using the four contacts low frequency lock-in technique with measuring currents lower than 100 μA. Experiments on $(TMTSF)_2PF_6$ were performed on a needle shaped crystal of dimensions (3 x 0.2 x 0.1) mm^3. As far as the $(TMTTF)_2BF_4$ salt is concerned, a first sample provided the 20 and 27 kbar resistivity data. A second one, of dimensions (1.5 x 0.075 x 0.05) mm^3 (along *a*, *b'*, *c** respectively), provided all the higher pressure data

3. RESULTS AND DISCUSSION

In the $(TMTSF)_2PF_6$ salt[5], measurements were conducted in 19 consecutive runs covering the 6.8-11kbar pressure range. Resistivity data for eleven different pressures are shown in Fig.1a. The resistivity curves present a strong increase in the residual resistivity as the pressure is lowered. However, a clear superconducting transition is observed with an onset critical temperature, T_c=1.20±0.01K, which is pressure independent. The resulting phase diagram is shown in Fig1.b.

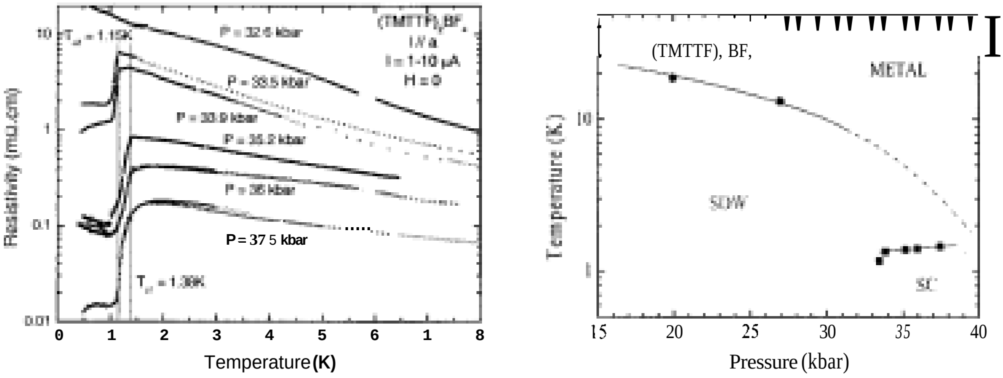


a) b)

Figure 1. a) Arrhenius plot of the resistance of $(TMTSF)_2PF_6$ versus temperature for different applied pressures. b) Pressure-temperature phase diagram of $(TMTSF)_2PF_6$ in the 8-11kbar range. SDW/M denotes the region where metallic, M, and SDW phases coexist inhomogeneously, below the T_{SDW} line (large full points). Below theT_{SC}=1.20±0.01K line, this coexistence switches into a coexistence of SC and SDW phases. In inset, the full diagram of $(TMTSF)_2PF_6$

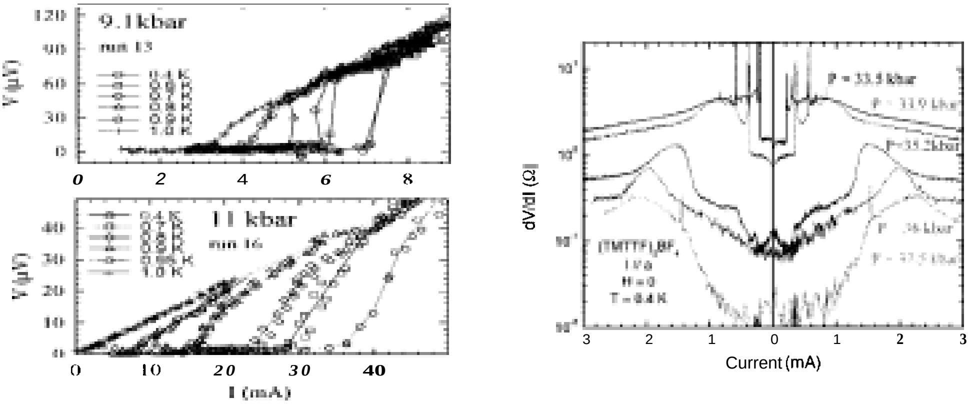
On the other hand, a superconducting state has been stabilized recently in $(TMTTF)_2$ PF_6, under 45 kbar [6] while a spin-Peierls ground state is present at ambient pressure. This transport experiment was performed in a Bridgman anvil cell up to 80 kbar with a solid pressure transmitting medium. Although the pressure range is limited to 40 kbar, the clamped cell technique provides hydrostatic pressure which is important for brittle organic samples, and allows easier measurements under magnetic field. We have chosen a TMTTF salt with a slightly smaller anion, BF_4, which is located on the right of the PF_6 salt in the generic phase diagram in order to get a lower critical pressure for SC and study the border between SDW and SC phases.

In this compound, at intermediate pressures, anion ordering induced Metal-Insulator transition is removed and a metallic behavior is preserved down to the SDW transition that we have observed at T_{SDW} =19 K for P = 20 kbar and 13 K for P = 27 kbar. These results allow us to extrapolate the critical pressure for the occurrence of SC to an intermediate value between the critical pressures of $(TMTTF)_2Br$ (26 kbar [7]) and $(TMTTF)_2PF_6$ (45 kbar [6]) as expected from an anion size argument. Indeed, superconductivity is reported in $(TMTTF)_2BF_4$ with $T_{SC} \approx 1.15$-1.38 K (Fig.2a) for pressures larger than 33.5kbar. However, localization appears prior to this transition implying that all investigated pressures are in the reentrant SC phase inside the SDW phase. We have performed successively on the same sample of $(TMTTF)_2BF_4$, six thermal cyclings corresponding to six different applied pressures from 37.5 down to 32.6 kbar in pressure steps between 0.5 and 1.5 kbar. The corresponding resistivity data presented on Fig.2a, show, when decreasing pressure, the gradual increase of the localization above T_{SC} and of the residual resistivity at zero temperature while T_{SC} remains constant in most of the coexistence region. It is only in the last kbar before the disappearance of SC, between 33 and 34 kbar, that T_{SC} is weakly decreasing. No sign of superconductivity could be detected at 32.6 kbar.



a) b)

Figure 2. a) Resistivity versus temperature curves in (TMTTF)$_2$BF$_4$. b) Pressure-temperature phase diagram of (TMTTF)$_2$BF$_4$.



a) b)

Figure 3. a) V(I) characteristics in (TMTSF)$_2$PF$_6$ for two different pressures. **b)** dV/dI(I) characteristics in (TMTTF)$_2$BF$_4$ for different pressures.

On the other hand, we performed non linear transport measurements : voltage-current characteristics performed on (TMTSF)$_2$PF$_6$ are shown in Fig.3a while dV/dI characteristics are shown for (TMTTF)$_2$BF$_4$ on Fig.3b for different applied pressures and at zero magnetic field. For both compounds, similar features are observed : the critical current decreases when applied pressure is lowered. In (TMTTF)$_2$BF$_4$, the SC critical current, I_c was taken at the first maximum of dV/dI (I). **As** the density of critical current for a fixed T_{SC} must be pressure independent in an homogeneous superconductor, the evolution of the critical current with pressure can be attributed only to a modification of the section of the sample which superconducts. The larger, the critical current, the larger is the size (or the number) of the SC domains inside the SDW phase. This segregation picture is reinforced by the observed decrease of dV/dI above I_c which is the signature of the background SDW in the depinned regime. This regime is also visible above T_{SC} in the pure SDW phase. At 0.45 K, the extrapolation of the saturation of Ic(P) at high pressures gives a density of critical current in the homogeneous SC phase around 80 A/cm^2 for an applied pressure of 40 kbar. This value is similar to the observed value in the homogeneous superconducting phase of (TMTSF)$_2$PF$_6$: 200A/cm^2.

The most striking feature of the dV/dI characteristics, in (TMTTF)$_2$BF$_4$, is the observation at the lowest pressures of strong peaks in the differential resistance. The shape of dV / dI is similar to previous reports in Sn whiskers [9] or more recently in nanotubes [10]. This strongly suggests that the SC domains are filamentary at the lowest pressures with a cross-section, perpendicular to the TMTTF chains, smaller than the coherence length. This image is reinforced by the large value of the upper critical field obtained from resistivity versus magnetic field curves shown in Fig.4 for (TMTTF)$_2$BF$_4$. At 0.45K, the upper critical field is 1.5Tesla, which leads to an extrapolated value $H_{c2} \approx$ 2T at zero temperature. It should be noted that the upper critical field is also nearly pressure independent as shown in Fig.4. This value is quite large compared to the usual 0.1-0.2T values for homogeneous superconductors in the $(TMTSF)_2X$ family but is compatible with old observations of a strong enhancement of the upper critical field as pressure is decreased in $(TMTSF)_2AsF_6$[11]. We suggest that this enhancement is probably due to the penetration of the magnetic field in the insulating regions in a similar way to Josephson vortices[12] in lamellar superconductors when the magnetic field is applied parallel to the superconducting planes.

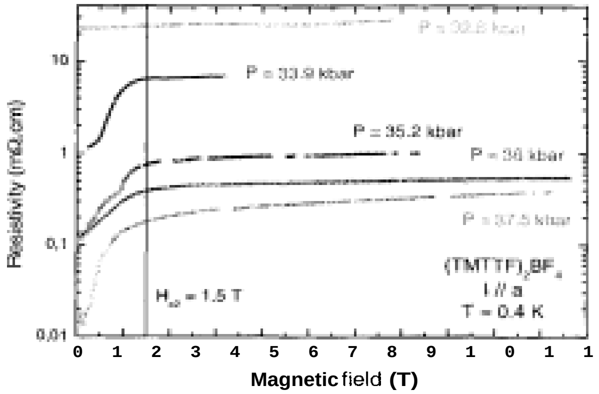


Figure 4. Resistivity versus magnetic field for H//c* in $(TMTTF)_2BF_4$.

4. CONCLUSION

We have demonstrated experimentally the coexistence of superconductivity and spin density wave orderings in both $(TMTSF)_2PF_6$ and $(TMTTF)_2BF_4$ in a narrow range of pressure : the critical temperature is nearly pressure independent, while the critical current collapses as pressure is decreasing. In $(TMTTF)_2BF_4$, we were able to show that the upper critical field is also pressure independent. All these features are compatible with a simple model of phase segregation we presented elsewhere [5].

Acknowledgements

The authors are grateful to M.Kończykowski (Ecole Polytechnique, Palaiseau) for providing the sensitive pressure gauge. The stay of T.Vuletić at LPS Orsay was in part supported by the project *Réseau formation recherche* of Ministry of Education and Research of the Republic of France.

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