THE ROLE OF CHARGE DENSITY WAVE FLUCTUATIONS IN STABILIZING THE GROUND STATE OF QUASI-TWO DIMENSIONAL CHARGE-TRANSFER SALTS, β”-(BEDT-TTF)$_4$[(H$_3$O)M(C$_2$O$_4$)$_3$]·Y

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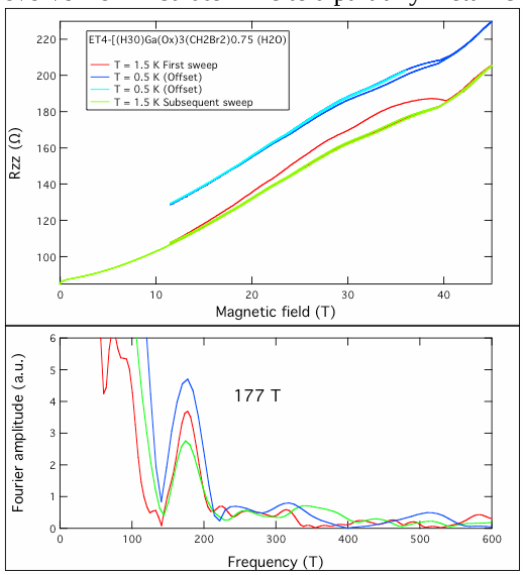
We have investigated the role of chemical pressure on the ground-state properties of the charge-transfer salts β”-(BEDT-TTF)$_4$[(H$_3$O)$\boldsymbol{M}$(C$_2$O$_4$)$_3$] · $\boldsymbol{Y}$ by varying the solvent molecule, $\boldsymbol{Y}$, from $\boldsymbol{Y}$ =C$_6$H$_5$NO$_2$ and C$_6$H$_5$CN (larger) to $\boldsymbol{Y}$ =C$_5$H$_5$N and CH$_2$Cl$_2$ (smaller). The transition-metal ion has also been varied from $\boldsymbol{M}$=Ga to $\boldsymbol{M}$=Cr and Fe. We have found that the size of the solvent is the main factor determining the nature of the ground state; this is either superconducting (for large solvent molecules) or a “bad metal” (for smaller solvent molecules). All salts show a metal-insulator transition associated with a charge-order-like transition [1] followed by a strong increase in resistance, as discussed in detail in Ref. [2]. Experiments in the 33 T and 45 T magnets at Tallahassee have shown that the Fermi surfaces of the superconducting and “bad metal” salts show important differences [1,2]. All samples exhibit Shubnikov-de Haas (SdH) oscillations superimposed on a negative or positive background magnetoresistance. Superconducting samples ($\boldsymbol{Y}$ =C$_6$H$_5$NO$_2$, C$_6$H$_5$CN) show two distinct SdH frequencies, whereas the metallic salts ($\boldsymbol{Y}$ =C$_5$H$_5$N and CH$_2$Cl$_2$) exhibit four frequencies. The presence of a series of pockets could be a consequence of a Fermi surface reconstruction at the metal-insulating transition; this may be more efficient in the superconducting salts. Similar Fermi-surface reconstructions have been suggested for other β”salts, including β”-(BEDT-TTF)$_2$AuBr$_2$ (where a plethora of small Fermi surface pockets results) and β”-(BEDO-TTF)$_2$ReO$_4$ ·H$_2$O [3,4]. Indeed, recent data on a new member of the β”-(BEDT-TTF)$_4$[(H$_3$O)$\boldsymbol{M}$(C$_2$O$_4$)$_3$] family (Figure 1) strongly reinforces both the presence of charge-density fluctuations and the similarity with β”-(BEDT-TTF)$_2$AuBr$_2$; hysteresis in the magnetoresistance indicates that the magnetic field is destabilizing a charge-density state. These findings suggest that the superconducting pairing in β”-(BEDT-TTF)$_4$[(H$_3$O)$\boldsymbol{M}$(C$_2$O$_4$)$_3$] · $\boldsymbol{Y}$ salts is probably mediated by change-density fluctuations, as predicted by Merino and McKenzie [5]. The phase diagram (Figure 1) shows that as the unit-cell size decreases the electronic properties of these salts evolve from insulator-like to a partially metallic character, with superconductivity adjacent to the charge-ordered state.



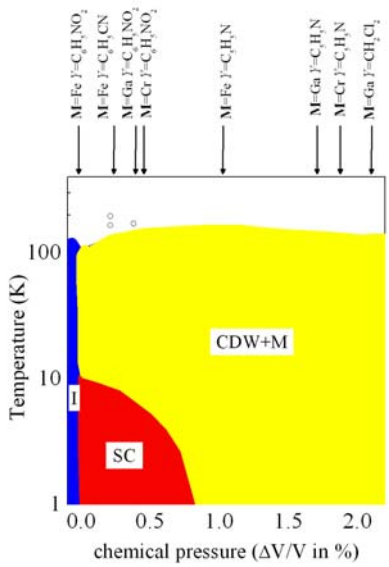


Figure 1. Left: the field dependence of the interplane resistance, R_{zz}, for the salt with M=Ga and Y=(CH$_2$Br$_2$)$_{0.75}$ H$_2$O at T = 0.5 and 1.5 K. The lower part of the figure is a Fourier transform of the same data showing the Shubnikov-de Haas frequency. Right: Notional phase diagram of the β”-(BEDT-TTF)$_4$[(H$_3$O)M(C$_2$O$_4$)$_3$] Y salts using data from Refs. 1 and 2.

References

[1] Bangura, A.F., *et al.*, Phys. Rev. B, **72**, 014543 (2005).
[2] Coldea, A.I., *et al.*, Phys. Rev. B, **69**, 085112 (2004).
[3] House, A.A., *et al.*, Phys. Rev. B, **53**, 9127 (1996).
[4] Kahlich, S., *et al.*, Z. Phys. B, **60**, 3060 (1999).
[5] Merino, J. and McKenzie, R.H., Phys. Rev. Lett., **87**, 237002 (2001).