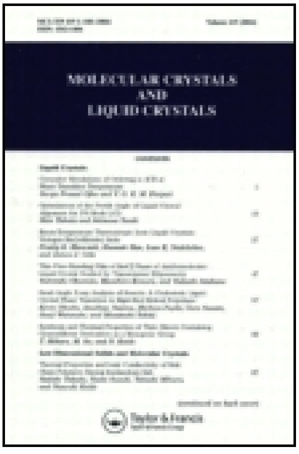

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

H-T Behavior of the Spin Density Wave Condensate in $(TMTSF)_2AsF_6$

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Version of record first published: 24 Sep 2006.

To cite this article: J. L. Musfeldt , M. Poirier , P. Batail & C. Lenoir (1996): H-T Behavior of the Spin Density Wave Condensate in $(TMTSF)_2AsF_6$, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 284:1, 121-128

To link to this article: http://dx.doi.org/10.1080/10587259608037916

Mol. Cryst. Liq. Cryst., 1996, Vol. 284, pp. 121–128
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Abstract We present the 16.5 GHz microwave dielectric response of $(TMTSF)_2AsF_6$ single crystals as a function of temperature and applied magnetic field ($H \parallel c^*$). Following the evolution of small features in ϵ_1 and σ_1 with temperature and field, we have generated an H–T diagram. The phase boundaries in this diagram seem to define changes in condensate behavior. Based upon the magnetic field dependence of the 3.5 K boundary, we conclude that this transition is not strongly driven by Fermi surface nesting considerations.

INTRODUCTION

Studies of the TMTSF-based synthetic metals have attracted a great deal of attention recently.[1,2] (Here, TMTSF is tetramethyltetraselenafulvalene.) The salts with octahedral anions (such as PF_6 or AsF_6) and 2:1 stoichiometry are prototypes in this family of organic materials due to their low-temperature metallic behavior and subsequent formation of the spin-density-wave (SDW) ground state near 12 K. Despite much study, many aspects of the broken symmetry state, the magnetic driving forces of transition, and the dynamics of the SDW condensate are still under investigation.[3–5]

Interest has focussed on the low-energy dielectric behavior of $(TMTSF)_2X$ materials because, in the low-temperature non-metallic state, the collective mode of the SDW dominates the response in these systems. Numerous microwave studies[6-13] have amply demonstrated the rich dielectric structure present in the low energy regime, with more recent studies probing the frequency dependent behavior for clues into SDW condensate dynamics. NMR has also been a leading investigative tool for characterizing the broken symmetry ground state in TMTSF-based charge-transfer salts.[14-21] Pertinent to the results presented in this paper, relaxation rate instabilities have been observed near 12, 3.5, and 1.9 K, dividing the low-temperature phase diagram into three regimes, commonly labeled SDW 1, SDW 2, and SDW 3, respectively.[18,19] The 3.5 K boundary has recently been attributed to a slowing down of phason dynamics,[17] likely brought on by a glassy transition.[4,5] Numerous other characterization methods have detected anomalous behavior near 3.5 K.[3,4,22,23] Because these various structures are observed in a similar temperature range, they are thought to be related. In this paper, we present high precision microwave dielectric measurements on $(TMTSF)_2AsF_6$ which display a characteristic signature near 3.5 K as well. We attribute the dielectric signature to a fundamental change in the SDW condensate behavior at the 3.5 K boundary.

Despite active study during the past decade, many questions still exist regarding the microwave dielectric response of the Bechgaard salts in the low-temperature phase. Manipulation of the overall dimensionality (via applied pressure or magnetic field) and measurement of the resulting physical properties have aroused special interest. That the SDW ground state is in direct competition with superconductivity is particularly intriguing.[18,19] Thus, our investigations of the 16.5 GHz microwave dielectric response of $(TMTSF)_2AsF_6$ as a function of temperature and applied magnetic field are directly pertinent to the on-going and overall inquiry into magnetic ground state properties in synthetic metal materials. Part of this work has been published elsewhere[24–26]

EXPERIMENTAL

Single crystals of $(TMTSF)_2AsF_6$ were grown by standard electrochemical methods. Our method of sample handling and for fixing the specimen in the intended orientation has been described previously.[24]

We have used a conventional cavity perturbation technique to measure the 16.5 GHz complex dielectric response $\epsilon^* = \epsilon_1 + i\epsilon_2$ of a $(TMTSF)_2AsF_6$ single crystal as a function of temperature and external magnetic field; here, ϵ_1 and ϵ_2 are the dielectric constant and the dielectric loss, respectively. ϵ_2 is related to the conductivity as $\sigma_1/\epsilon_0\omega$. Thus, we measure both ϵ_1 and σ_1 at the resonance frequency of the cavity. A detailed description of our experimental set–up is given elsewhere.[24] We have made both temperature and magnetic field sweeps of the measured parameters, concentrating on the 1.7-7 K and 0–3 T range. A few runs were made to higher temperature and field. The results were analysed within the framework of the quasi–static approximation,[27] allowing us to calculate the real and imaginary parts of the dielectric constant at the probe frequency. Our rational for pursuing an analysis based upon the quasi–static approximation is detailed in Ref. 24. We identified and followed progressive changes in ϵ_1 and σ_1 which appear as a function of temperature and applied magnetic field. From these data, we have constructed an H–T diagram. For a detailed discussion of our derivative method for identifying the phase boundaries and our estimate of the various errors involved, please refer to Refs. 25, 26.

RESULTS AND DISCUSSION

Zero Field Response

Figure 1 displays the zero field 16.5 GHz dielectric constant of $(TMTSF)_2AsF_6$, together with the temperature derivative of ϵ_1. The value of ϵ_1 is large, consistent with the highly polarizable nature of the condensate at microwave frequencies and in good agreement with previous estimates of ϵ_1 in $(TMTSF)_2X$-type materials.[4,9,13] Furthermore, the overall shape of ϵ_1 is consistent with that obtained on the PF_6 sample in a similar experiment.[24]

The abrupt change in ϵ_1 through the 12 K metal $\rightarrow$ insulator transition suggests that quasi-particle effects dominate the dielectric response both near T_c and in the higher temperature regime. In contrast, the 3.5 K drop in ϵ_1 (and the corresponding local maximum in σ_1) is probably unrelated to quasi-particle effects, as it occurs far from T_c where the quasi-particle effects are nearly frozen out and the DC conductivity is featureless.[22] Thus, we conclude that the 3.5 K structure is a signature of the changing condensate behavior at the glassy (SDW 1 $\rightarrow$ SDW 2) transition. The 1.85 K upturn in ϵ_1 is likely related to changing condensate behavior as well.

Magnetic Field Effects

The effect of applied magnetic field ($H \parallel c^*$) on the dielectric constant is displayed in Fig. 2. The confinement induced by the applied field produces strong changes in the 3.5 and 1.9 K dielectric features. We have mapped the location of these changes (for both field and temperature sweeps) to obtain the H–T diagram described below.

The H–T diagram for the condensate behavior of $(TMTSF)_2AsF_6$ at 16.5 GHz ($H \parallel c^*$) is shown in Fig. 3. We have identified the "zero field boundaries" at $\approx$ 12.1 (not shown), 3.4, and 1.85 K from aforementioned dielectric features on

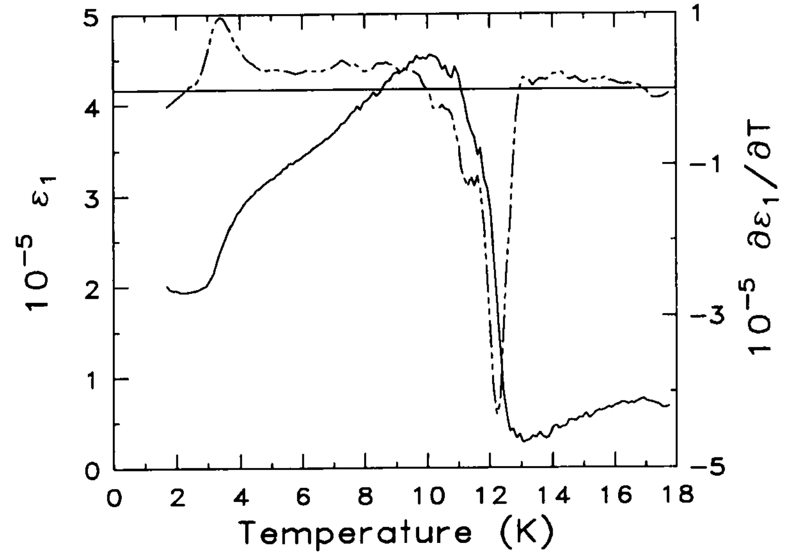

Figure 1: Solid line: 16.5 GHz dielectric constant of $(TMTSF)_2AsF_6$ as a function of temperature taken at zero magnetic field; double-dashed line: $\partial\epsilon_1/\partial T$ of the zero field curve, illustrating how points in Fig. 3 were determined.

the zero-field curve. These values are in excellent agreement with the anomalies observed in lattice specific heat measurements.[3,4] The boundaries defined by these points move to higher temperature and broaden slightly with increasing magnetic field. Thus, SDW 2 (and to a lesser extent, SDW 3) is stabilized at the expense of SDW. A similar magnetic field dependence for the 3.5 K boundary can be inferred from the NMR data of Clark *et. al.*[16]

The strong magnetic field dependence of the 3.5 K boundary is indicative of a change in dynamics in which Fermi surface nesting plays a minimal role. Indeed, these results and others point toward different driving forces at 12 K, where nesting considerations dominate,[28,29] *vs.* 3.5 K, where large condensate effects are driven by glassy dynamics.[4] The magnetic field dependence of the 3.5 K boundary is qualitatively consistent with a reduction of the SDW pinning length with applied field.[30]

In addition to resolving the magnetic field dependence of the aforementioned

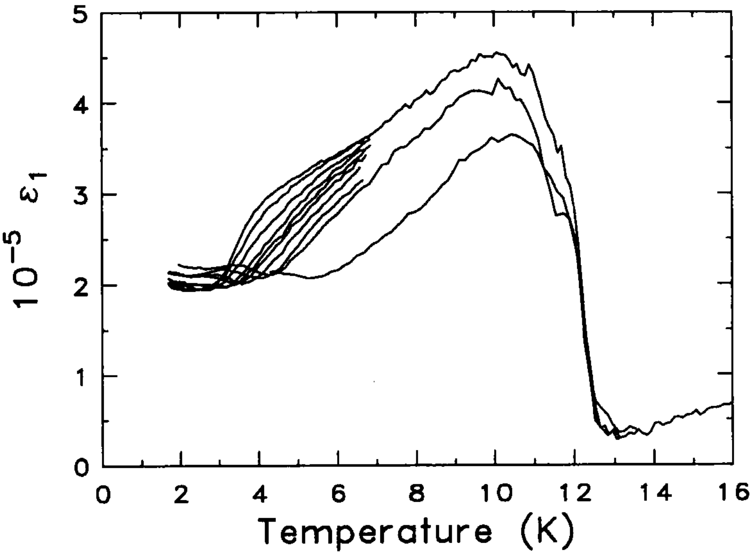

Figure 2: 16.5 GHz dielectric constant of $(TMTSF)_2AsF_6$ as a function of temperature taken at various applied magnetic fields (H = 0.2, 0.5, 0.8, 1.0, 1.4, 1.8, 2.5, 3.0, 7.0. Curves change monotonically with increasing H.

condensate boundaries, we observe preliminary evidence for a low-temperature/low-field region within SDW 2. This structure does not seem to be *directly* attributable to the spin-flop transition,[26,31] instead relating perhaps to sampling of off-diagonal components of $\epsilon_1{}^*$ or the nature of the glassy domain structure below 3.5 K.

CONCLUSION

We have reported the 16.5 GHz dielectric response of the low-dimensional Bechgaard salt $(TMTSF)_2AsF_6$ at temperatures above and below the 12 K antiferromagnetic phase transition. The zero field response is in good agreement with previous results obtained for the PF_6 salt. Field and temperature sweeps concentrating in the 1.7–7 K and 0–3 T range have allowed us to construct an H-T diagram, which illustrates the complex behavior of the condensate at low temperatures and applied fields ($H \parallel c^*$). More work is needed to fully characterize the

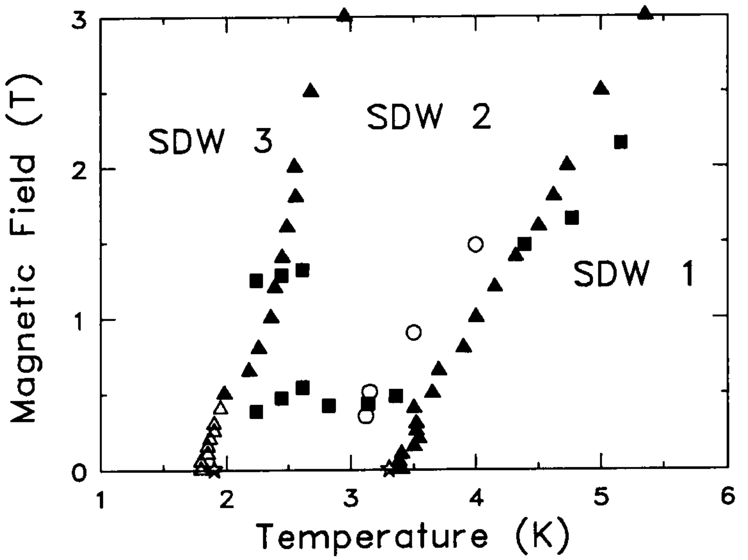

Figure 3: The H–T behavior of the SDW condensate of $(TMTSF)_2AsF_6$ at 16.5 GHz. The external magnetic field is applied along the hard axis (c^*) direction. Filled triangles: position of dielectric anomalies as determined by temperature sweeps at constant field; filled squares: position of dielectric anomalies as determined by magnetic field sweeps at constant temperature; open triangles: position of the low–temperature/low–field dielectric anomaly as *estimated* from temperature sweeps at constant fields; open circles: NMR data of Clark *et. al.* (Ref. 16); open stars: lattice specific heat data of Lasjaunias *et. al.* (Ref. 4). Note that the data in Refs. 16 and 4 were obtained on the PF_6 salt.

nature of the various low–temperature states in this interesting material.

ACKNOWLEDGEMENTS

This work was supported by grants from the Natural Science and Engineering Research Council of Canada (NSERC) and the Fonds pour la Formation de Chercheurs et l'Aide à la Recherche (FCAR) of the Government of Québec. We greatly appreciate the technical assistance of M. Castonguay and C.D. Porter, as well as helpful conversations with C. Bourbonnais, L. Chen, and Y. Trudeau.

REFERENCES

1. G. Grüner, Rev. Mod. Phys., 66, 1 (1994).
2. T. Ishiguro and K. Yamaji in Organic Superconductors, Springer Series of Solid State Sciences, Vol. 88, P. Fulde, ed. (Springer–Verlag, Berlin, 1990).
3. J.C. Lasjaunias *et. al.*, Sol. State. Commun., 84, 297 (1992).
4. J.C. Lasjaunias *et. al.*, Phys. Rev. Lett., 72, 1283 (1994).
5. J. Odin *et. al.*, Solid State Commun., 91, 523 (1994).
6. S. Donovan *et. al.*, Phys. Rev. B., 49, 3363 (1994).
7. S. Donovan *et. al.*, J. Phys. I France, 3, 1493 (1993).
8. S. Donovan *et. al.*, Euro. Phys. Lett., 19, 433 (1992).
9. G. Mihály *et al.*, Phys. Rev. Lett., 67, 2806 (1991).
10. D. Quinlivan *et. al.*, Phys. Rev. Lett., 65, 1816 (1990).
11. H.H.S. Javadi *et. al.*, Phys. Rev. Lett., 55, 1216 (1985).
12. A. Jánossy *et. al.*, Solid State Commun., 46, 21 (1983).
13. A. Zettl *et. al.*, Phys. Rev. B., 25, 1443 (1982).
14. W.H. Wong *et. al.*, Phys. Rev. Lett., 72, 2640 (1994).
15. E. Barthel *et. al.*, Euro. Phys. Lett., 21, 87 (1993).
16. W.G. Clark *et. al.*, J. Phys. IV, 3, 235 (1993).
17. W.H. Wong *et. al.*, Phys. Rev. Lett., 70, 1882 (1993).
18. T. Takahashi *et. al.*, Synth. Met., 41-43, 3985 (1991).
19. T. Takahashi *et. al.*, J. Phys. Soc. Jpn., 55, 1364 (1986).
20. C. Bourbonnais *et. al.*, Phys. Rev. B., 33, 7608 (1986).
21. J.M. Delrieu *et. al.* J. Phys., 47, 839 (1986).
22. G. Kriza *et. al.*, Europhys. Lett., 16, 585 (1991).
23. J.P. Ulmet *et. al.*, Synth. Met., 19, 271 (1987).
24. J.L. Musfeldt *et. al.*, Phys. Rev. B., 51, 8347 (1995).
25. J.L. Musfeldt *et. al.*, Euro. Phys. Lett., 30, 105 (1995).
26. J.L. Musfeldt *et. al.*, Phys. Rev. B., *accepted.*
27. L.I. Buranov and I.F. Shchegolev, Instrum. and Exp. Tech., 14, 528 (1971).
28. K. Yamaji, J. Phys. Soc. Jpn., 51, 2787 (1982).
29. G. Montambaux, Phys. Rev. B., 38, 4788 (1988).
30. A. Bjeliš and K. Maki, Phys. Rev. B., 44, 6799 (1991).
31. K. Mortensen *et. al.*, Phys. Rev. B., 25, 3319 (1982).