Collective spin-density-wave response perpendicular to the chains of the quasi-one-dimensional conductor $(TMTSF)_2PF_6$

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Microwave experiments along all three directions of the spin-density-wave model compound $(TMTSF)_2PF_6$ reveal that the pinned mode resonance is present along the a and b' axes. The collective transport is considered to be the fingerprint of the condensate. In contrast to common quasi-one-dimensional models, the density wave also slides in the perpendicular b' direction. The collective response is absent along the least conducting c^* direction.

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The electrodynamic response of quasi-one-dimensional materials with a density-wave ground state has been thoroughly explored during past decades. At low temperatures the optical conductivity develops an absorption edge in the infrared spectral range due to the opening of the single-particle gap at the Fermi energy. A phason mode (so-called pinned mode) is usually found in the GHz range of frequency; it can be attributed to the collective response of the condensate pinned to lattice imperfections. At even lower frequencies (in the range of MHz, kHz and even below, depending on temperature) internal deformations and screening by the conduction electrons lead to a broad relaxational behavior. Numerous experimental and theoretical studies performed on model compounds for the formation of charge density waves (CDW), like TTF-TCNQ, $K_{0.3}MoO_3$, $NbSe_3$, or $(TaSe_4)_2I$, and the formation of spin-density waves (SDW), like $(TMTSF)_2PF_6$, have been summarized in a number of reviews and monographs.[1–3] The generic conductivity spectrum is plotted in Fig. 1 for the example of $(TMTSF)_2PF_6$.

Basically all the experiments and models focus on the transport along the highly conducting chains. However, real materials are three-dimensional: no matter how anisotropic they are, the interaction between the chains cannot be neglected. In fact it is known that the three-dimensional coupling between the density wave formed on the chains is essential to develop the ordered ground state at finite temperatures.[5,6] Some of the most studied density-wave systems, $K_{0.3}MoO_3$ and $(TMTSF)_2PF_6$, in fact have a tendency toward two-dimensionality; for our example of $(TMTSF)_2PF_6$ the transfer integrals $(4t_a:4t_b:4t_c) = (1.5:0.1:0.003)$ eV have been determined from band structure calculations.[7] In certain cases the dc transport was measured for the perpendicular directions. Similar to the resistivity along the highly-conducting axis, the density-wave transition can in general also be observed by a sharp increase of the resistivity perpendicular to the chains (cf. Fig. 2). The explanation is the opening of the single-particle gap over the entire Fermi surface, or at least large parts of it. Very little, however, is known about the collective response which (besides sophisticated methods like narrow-band noise) can best be observed by a threshold field in the nonlinear conductivity or by the pinned-mode resonance. It has been argued that the density wave is a strictly one-dimensional phenomenon which develops only along the chains.[8] The aim of this study is the search for indications of the collective electrodynamic response of the SDW in the perpendicular directions.

Single crystals of the Bechgaard salt di-(tetramethyltetraselenafulvalene)-hexaflourophosphate, denoted as $(TMTSF)_2PF_6$, were grown by electrochemical methods.[9] The properties of this family of organic conductors has been summarized by Jérome.[10] The dc resistivity $\rho(T)$ of $(TMTSF)_2PF_6$ along the a axis was measured on needle-shaped samples with a typical dimension of $(2\times0.5\times0.1)$ mm^3 along the a, b', and c^* axes, respectively. Due the triclinic symmetry, b' denotes the projection of the b axis perpendicular to a, and c^* is normal to the ab plane. The b'-axis conductivity was obtained on a narrow slice cut from a thick crystal perpendicular to the needle axis; the typical dimensions of so-made samples were $a\times b'\times c^* = (0.2\times1.3\times0.3)$ mm^3. Due to the large sample geometry, the b'-axis resistivity was measured for the first time with basically no influence of the a and c^* contributions and using standard four-probe technique to eliminate the contact resistances. Also for the c^*-axis transport, four contacts were applied, two on each side of the crystal. In addition, the microwave conductivity at 24 and 33.5 GHz was measured in all

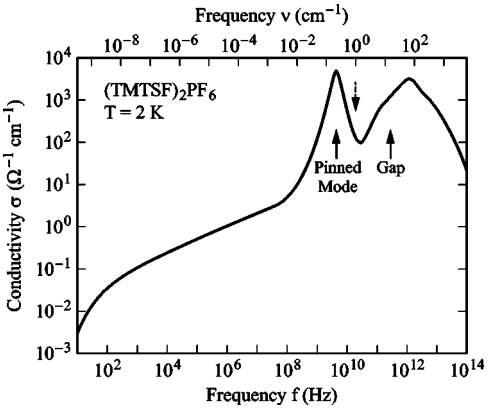

FIG. 1. Sketch of the frequency-dependent SDW conductivity composed using data along the chain axis of $(TMTSF)_2PF_6$. (Ref. 4). The solid arrows indicate the position of single particle gap and the pinned mode resonance in the microwave frequency range. The dashed arrow depicts the frequency range of the investigations presented in this work.

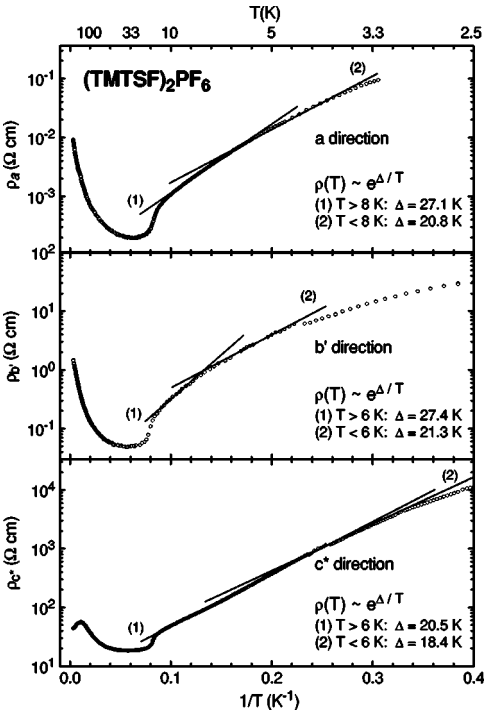

FIG. 2. Arrhenius plot of the temperature dependent dc resistivity of $(TMTSF)_2PF_6$ along the a, b', and c^* directions.

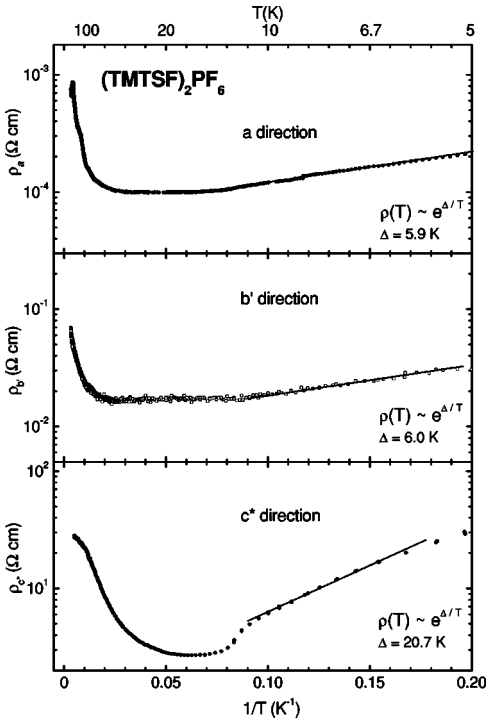

FIG. 3. Temperature dependent microwave resistivity of $(TMTSF)_2PF_6$ along the a, b', and c^* directions measured at 33.5 GHz.

three directions. The crystals were placed onto a quartz substrate and positioned in the maximum of the electric field of a cylindrical copper cavity. Along the a direction the naturally grown needles were used, because this geometry is best for precise microwave measurements. As described above, a slice was cut from a thick single crystal to measure in b' direction. In order to perform microwave experiments along the c^* axis, a crystal was chopped into several pieces (approximately cubes of 0.2 mm corner size) and arranged up to four as a mosaic in such a way that a needle-shaped sample of about $(0.2\times0.2\times0.8)$ mm^3 was obtained. By recording the center frequency and the halfwidth of the resonance curve as a function of temperature and comparing them to the corresponding parameters of an empty cavity, the complex electrodynamic properties of the sample, like the conductivity and the dielectric constant, can be determined via cavity perturbation theory; further details on microwave measurements and the data analysis are given in Refs. 9 and 11.

In Fig. 2 the temperature dependence of the dc resistivity is plotted. When the SDW ground state develops at $T_{SDW}=12$ K a sharp increase of $\rho(T)$ is observed along the a, b', and c^* directions. For $T<T_{SDW}$ an activated behavior $\rho(T)\propto\exp\{\Delta/T\}$ can be identified, with a single-particle energy gap of 27.1 K, 27.4 K, and 20.5 K along a, b', and c^* directions, respectively. On cooling down further (somewhat below 6 K) the activation energy is slightly reduced giving values of 20.8 K, 21.3 K, and 18.4 K in the three orientations. At very low temperatures a saturation of $\rho(T)$ is observed since a finite number of metallic carriers remains due to the nonperfect nesting.[1] While for the a and b' axes the activation energy is identical within the error bars, a somewhat lower value is observed for the least conducting direction. These data are in good agreement with earlier findings[12–15] and estimations by mean field theory: $\Delta(T=0)=3.53T_{SDW}/2\approx21$ K.

Also in the microwave data, the SDW transition at 12 K is present in all three directions. The temperature-dependent resistivity measured at 33.5 GHz is plotted in Fig. 3 in the Arrhenius representation. Up to six samples of different batches have been studied for each orientation; the sample-to-sample spread is within the uncertainty to determine the slope. Similar results are obtained at 24 GHz, but both frequencies are too close to allow for any conclusions on the frequency dependence. Most surprisingly, the activation energy along the a and b' axes is much smaller compared to the dc behavior, while for the c^* orientation the results at microwave frequencies perfectly agree with the dc profile. Right below T_{SDW} the activation energies obtained for the three directions are (5.9 ± 0.4) K, (6.0 ± 0.3) K, and (20.7 ± 0.4) K. The significantly reduced values of the activation energy for the a and b' directions compared to dc data infer a strong frequency dependent response which is associated with the collective mode contribution to the electrical transport.

Based on an extensive microwave study along the chain direction, it was proposed[4] that due to impurity pinning the collective SDW response in $(TMTSF)_2PF_6$ is located around 5 GHz. The conductivity below the energy gap decreases exponentially with decreasing temperature, except in the range of the pinned mode. As can be seen from Fig. 1, the present microwave experiments are performed in the range where the collective mode is still very pronounced, i.e., on

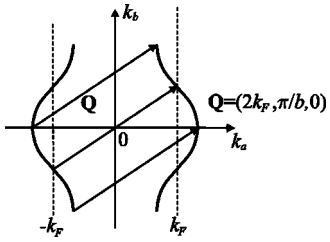

FIG. 4. Schematic Fermi surface nesting of a quasi-one-dimensional system with interchain coupling in the *b* direction.

the shoulder of the pinned mode resonance. Hence the temperature dependent microwave conductivity is caused by two opposing effects: (i) the exponential freeze-out of the background conductivity caused by the uncondensed conduction electrons, and (ii) the build-up of the collective contribution. It was suggested that this mode does not gain much spectral weight as the temperature decreases, but the width and center frequency changes slightly.[4] The most surprising discovery of our investigation is the presence of the enhanced microwave conductivity not only along the chains, but also perpendicular to them. This implies that the pinned mode resonance is present in the b' direction in a very similar manner compared to the *a* axis. Our conclusions are supported by previous studies of Balicas[16] on the nonlinear transport in the field-induced spin-density-wave phases ($p=8.5$ kbar, $B=7.23$ T, $T=0.37$ K) which indicate that the phason mode is responsible for the break down of the quantum-Hall-effect regime. Current models assume that the density wave can slide only along the chain direction;[8] our findings, however, give clear evidence for a collective contribution to the conductivity in the perpendicular direction. No indications of a collective response is observed along the c^* direction. Hence the sliding density wave has to be considered a two-dimensional phenomenon with severe implications on the theoretical description.

These results can be explained by looking at the actual Fermi surface of $(TMTSF)_2$-PF_6 which is not strictly one dimensional but shows a warping in the direction of k_b (and much less in k_c), as depicted in Fig. 4. From NMR experiments[17] it is known that the SDW corresponds to a wave vector $\mathbf{Q}=[0.5a^*,(0.24\pm0.03)b^*,(-0.06\pm0.20)c^*]$; which is incommensurate with the underlying lattice. Most important in this context, is an appreciable component of **Q** in the *b* axis. The tilt of the nesting vector is responsible for the similar collective SDW response found in the microwave experiments along the *a* and b' directions.

In a number of papers, Maki and Virosztek studied the collective transport of SDW in organic conductors; in particular they considered charge- and spin-density-wave systems in the presence of long-range Coulomb interaction and found significant differences if the nesting vector deviates from the chain direction.[18,19] More detailed investigations, however, are needed to calculate and also measure the influence of pinning, the nonlinear conduction, and the spectral weight of the collective mode perpendicular to the chains.

In conclusion, the enhanced conductivity found by microwave experiments on $(TMTSF)_2$-PF_6 evidences a collective transport not only along the chains, but also in the perpendicular b' direction. In contrast to the present view, the sliding SDW condensate is not confined to the chains but it is a two-dimensional phenomenon. This conclusion, in fact, should hold for most of the spin and charge density wave materials. Similar investigations (including studies of the *I-V* characteristic) on the quasi-one-dimensional CDW model compound $K_{0.3}MoO_3$ are in progress.

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