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Short Communication

Motional narrowing of the nuclear magnetic resonance line by the sliding of spin-density waves

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Abstract. — We report on a study of the electrical conductivity and ^{13}C-NMR in the nonlinear
conductivity regime in the spin-density wave (SDW) phase of $(TMTSF)_2PF_6$ (TMTSF stands
for tetramethyltetraselenafulvalene). The narrowing of the NMR line demonstrates that there
is a bulk motion of the SDW. We find a qualitative agreement between the nonlinear current
and the minimum current necessary to narrow the NMR line in the simplest model of SDW
conduction.

The anisotropic organic conductor $(TMTSF)_2PF_6$ [1] is in several respect the prototype material for the study of the properties of the one dimensional (1-D) electron gas. A very striking signature of 1-D physics is the competition between electron-electron and electron-hole pairing instabilities.

Due to the stœchiometry of the salt and its crystalline structure, $(TMTSF)_2PF_6$ is formally a half-filled band 1-D conductor. This system is sensitive to electronic instabilities. At ambient pressure, the most stable state is a ground state exhibiting a finite modulation of the magnetization or spin-density wave (SDW) [2]. The divergence towards the SDW state is dominant as long as the interchain coupling remains sufficiently small. Hence, the application of a high pressure, which enhances the transverse coupling makes superconductivity the most stable state at 9 kbar [3].

The SDW state is phenomenologically antiferromagnetic, as demonstrated by susceptibility anisotropy [4] data and antiferromagnetic resonance [5]. The main difference between a regular antiferromagnetic phase and the SDW phase lies in the wave vector of the modulation. It was clearly shown from the analysis of μSR [6] and proton NMR [7, 8] spectra that the wave vector in $(TMTSF)_2PF_6$ is incommensurate with the lattice.

One of the most spectacular properties of this compound is the sharp increase of conductivity above a low threshold field [9]. It is widely believed that the incommensurate character of the

SDW ground state is at the origin of a zero frequency collective mode, which in nonlinear response amounts to a sliding of the overall electron-hole condensate. Pinning of the SDW to impurities accounts for the nonzero threshold field.

The same nonlinear conductivity is found in charge-density waves (CDW). In the NMR spectrum, the presence of the CDW induces a broad frequency distribution because of the spatial distribution of the electric field gradient. In the nonlinear conductivity regime, this line is narrowed [10]. This clearly demonstrates that the additional conductivity induces a fast modulation of the local field, thus supporting the notion of sliding.

A ^{1}H-NMR study of the sliding SDW in the quenched SDW state of $(TMTSF)_2ClO_4$ has been reported recently [11]. However, the protons are dipolarly coupled in four distinct methyl groups, and give rise to a very complex structure. The ^{13}C-NMR at the carbon sites at the center of the TMTSF molecule are much better suited for these studies, because the spectrum is simpler, and the coupling of these nuclei to the SDW is stronger than the coupling of protons. We demonstrated in a previous paper [12] that the ^{13}C-NMR line in $(TMTSF)_2PF_6$ is also considerably broadened in the SDW state.

In this communication, we report the preliminary results of a study of the electrical conductivity and ^{13}C-NMR in the nonlinear conductivity regime of $(TMTSF)_2PF_6$. Narrowing of the NMR line demonstrates that there is a bulk motion of the SDW. We find a qualitative agreement between the injected current and the minimum current necessary to narrow the line in the most simple model of sliding.

The sample comes from the same batch of selectively ^{13}C enriched crystals [13] we used in our previous measurements [12]. The occupation probability for each of the central sites (labelled (3) and (13) in [14]) is 5 %. Its size was $3.0 \times 0.25 \times 0.3$ mm^3, with a cross-section of 6×10^{-4} mm^2 and a mass of 0.35 mg. All measurements were performed in the 9.3 T NMR field. For the contacts, we did not use silver paint, because it is notorious for cracking upon cooldown, but two clamped contacts applied on the gold-plated b-c faces of the sample. No jump in the resistivity was observed upon cooldown, and the residual resistivity at 15 K was 0.9 Ω. The 18 Ω resistance at 4.2 K is thus almost entirely due to the sample, in agreement with the very large transverse magnetoresistance of the SDW phase [15].

At 4.2 K, in the SDW phase, we clearly observe a threshold field of 5 mV/cm in the current-voltage $(I$-$V)$ characteristics (Fig. 1), which is consistent with the usual values [16]. We obtained the NMR spectra (Fig. 2) by an echo sequence with a Bruker MSL400 spectrometer. At 9.3 T, the ^{13}C frequency is 99.67 MHz. At zero current, the spectrum exhibits two very wide U-shaped structures which are typical of the inhomogeneous broadening due to the sinusoidally spatially modulated magnetization. Each one is associated with one of the two inequivalent sites. The contribution of the other carbons is negligible because of the small ^{13}C concentration and long relaxation time.

Under current, two narrow peaks grow out of the broad distributions. Their intensity increases with increasing current. However, even at 1 mA, there remains a reminiscence of the zero-current spectrum. We believe this is due to the inhomogeneity of the current density in the sample. Therefore, there is a substantial fraction of the sample in which no SDW current flows. We estimate this fraction as 75 % at 0.5 mA, and as 60 % at 1 mA.

At the same time, the presence of a static fraction clearly demonstrates that the appearance of the two peaks in the nonlinear conductivity regime is not due to heating of the sample above the phase transition. We also stress that if heating were involved, the intensity of the NMR signal should decrease by at least a factor of 3, which is not observed.

These results confirm the sliding SDW picture. However, the exact nature of the sliding mode can be characterized further by the ratio of the SDW current per chain, j_{SDW}, to the local phase oscillation frequency ν_ϕ (phase winding rate). This ratio, in the simplest model
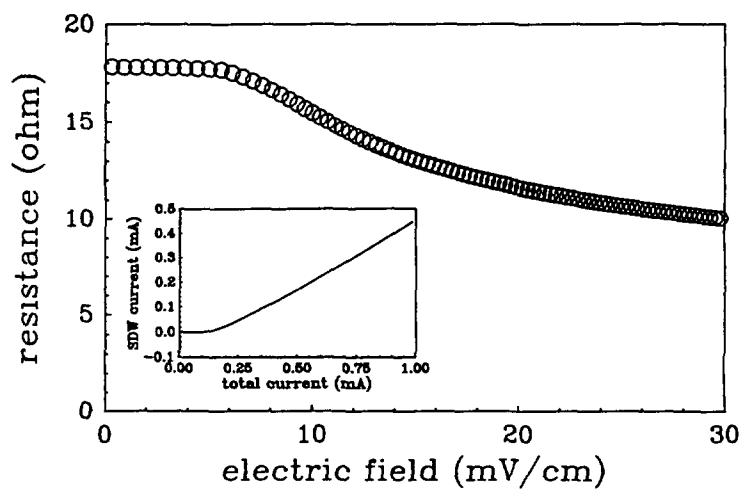
Fig.1. — Sample resistance as a funtion of electric field ($T = 4.2$ K). Inset: nonlinear SDW current $I_{SDW} = I - V/R(E \rightarrow 0)$ as a function of the total current I.
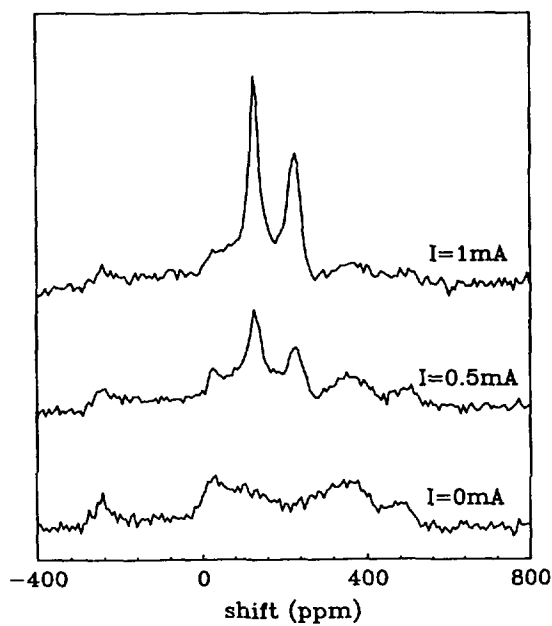

Fig.2. — NMR spectra in the SDW phase as a function of the total sample current I at $T = 4.2$ K.

[17], is expected to be:

$$j_{\text{SDW}}/\nu_\phi = 2e \,. \tag{1}$$

It can be indirectly determined by measuring the frequency of the conduction noise which appears in the nonlinear conductivity regime *versus* current. Up to now, these determinations [18] have yielded a ratio which is between 8 and 20 times smaller than expected from equation (1). Although it is not clear whether the noise frequency is equal to the phase winding rate or twice larger, these data suggested a large discrepancy with the theory. It is usually assumed that this arises as a strongly inhomogeneous depinning, which leads to an erroneous estimate of the effective sliding cross-section.

We estimate the SDW current from

$$I_{\text{SDW}} = I - V/R \,,$$

where R is the low electric field resistance (Fig. 1, inset). Then, using a chain cross section of 100 Å^2 and a depinned fraction inferred from the NMR spectra, we find from equation (1) a mean phase winding rate of 30 kHz at a total current of 0.5 mA, and 56 kHz at 1 mA. The simulations show that such frequencies are high enough to yield a substantial narrowing of the line. Therefore, we cannot exclude, on the basis of these data, that the j_{SDW}/ν_ϕ ratio is considerably smaller than expected. However, this is not necessary.

We conclude that there are large amplitude local phase fluctuations in the non linear conductivity regime in $(\text{TMTSF})_2\text{PF}_6$. This result is consistent with the accepted picture of a sliding spin-density wave. Although a small j_{SDW}/ν_ϕ ratio is not in contradiction with our results, it is not necessary to explain our data. Obviously, combined conduction noise and NMR measurements are needed to solve this problem.

After this work was performed we learned that W. H. Wong and coworkers [19] had also observed sliding SDW in $(\text{TMTSF})_2\text{PF}_6$ by ^{1}H-NMR in low magnetic fields.

Acknowledgments.

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Corrigendum and addendum to section 3 of "Level spacing functions and non linear differential equations"

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In the above reference we missed to note that the functions $\mathcal{A}$, w, u, v, and $i\xi$ in equations (3.11)–(3.21) are real for real τ, so that the absolute value of $y_{a2}(\tau)$ is unity for real τ. In other words, if we set $y_{a2} = \exp(2i\rho)$, $\rho(\tau)$ is real for real τ, satisfies the (non linear) differential equation

$$\left(\rho'' + \frac{\rho' + 2}{\tau}\right)\tan\rho - \rho'^2 + 4 = 0 \tag{3.28}$$

and near $\tau = 0$ has the expansion

$$\rho(\tau) = -2\tau\left[1 - \zeta\tau + O(\tau^3)\right] \tag{3.29}$$

Thus equation (3.5) should be corrected as

$$
\begin{aligned}
y_{a2} &= \exp\left[-4i\tau + 4i\zeta\tau^2 + O(\tau^4)\right] \\
&= 1 - 4i\tau + 4(i\zeta - 2)\tau^2 + 16\left(\zeta - \frac{2i}{3}\right)\tau^3 + O(\tau^4).
\end{aligned} \tag{3.5'}
$$

Also it was shown that each of the four functions Re $\mathcal{S}(\tau)$, Im $\mathcal{S}(\tau)$, $\mathcal{A}(\tau)$, $\mathcal{B}(\tau)$ can be expressed in terms of a fifth Painlevé transcendent (P5), and one of them, namely $\mathcal{B}(\tau)$, also in terms of a third Painlevé transcendent (P3). Actually, according to Gromak (see the reference below), each of the remaining three functions can also be expressed in terms of a P3. In other words, as the P5 with equation (3.5') is related to the P3 with equation (3.8), so the P5 with equations (3.2), (3.3) and (3.4) are related to the P3 with respectively

(1) $\alpha = -1$, $\beta = 3$, $\gamma = 1$, $\delta = -1$; and for $\tau \ll 1$,

$$y(\tau) = -\frac{1}{\zeta} + \left(3 - \frac{1}{\zeta^2}\right)\tau + \left(-\frac{1}{\zeta^3} + \frac{3}{\zeta} - 2\zeta\right)\tau^2 + \quad ; \tag{3.30}$$

(2) $\alpha = -1$, $\beta = 3$, $\gamma = 1$, $\delta = -1$; and for $\tau \ll 1$,

$$y(\tau) = \frac{\tau}{3} + \frac{\tau^3}{45} - \frac{\zeta\tau^4}{27} + \cdot \quad ; \tag{3.31}$$

and
(3) $\alpha = 1$, $\beta = 1$, $\gamma = 1$, $\delta = -1$; and for $\tau \ll 1$,

$$y_+(\tau) = -\tau + \zeta\tau^2 - \frac{\tau^3}{3} + \cdot \quad , \tag{3.32}$$

or another possible choice

$$y_-(\tau) = -\frac{1}{\tau} - \zeta - \left(\zeta^2 - \frac{1}{3}\right)\tau + \cdot \tag{3.33}$$

Here are the necessary details. This addendum is supposed to be read with the original reference.

Consider the couple of equations

$$f = \frac{\sqrt{\tau}}{R^2 - \tau}\left(R' - \frac{R}{\tau}\right), \qquad R = -\frac{\sqrt{\tau}}{f^2 - 1}\left(f' + \frac{3}{2}\frac{f}{\tau}\right). \tag{3.34}$$

Eliminating f one gets equation (3.10), while eliminating R one gets

$$f'' = \frac{f}{f^2 - 1}f'^2 - \frac{f'}{\tau} - \frac{9}{4\tau^2}\frac{f}{f^2 - 1} + f(f^2 - 1). \tag{3.35}$$

Set $f = \sqrt{u/(u-1)}$, or $u = f^2/(f^2 - 1)$, so that

$$u'' = \frac{3u - 1}{2u(u-1)}u'^2 - \frac{u'}{\tau} + \frac{9u}{2\tau^2}(u-1)^2 - 2u. \tag{3.36}$$

This is again almost a P5 with $\delta = 0$, and becomes a standard P5 on taking τ^2 as the new independent variable.

Consider now the couple of equations

$$\frac{u+1}{u-1} = \omega^2 - \omega' - \frac{2\omega}{\tau}, \qquad \omega = \frac{2\tau u}{\tau u' - 3u(u-1)}. \tag{3.37}$$

Eliminating ω, one gets (3.36), while eliminating u one gets a P3 for ω with $\alpha = -1$, $\beta = 3$, $\gamma = 1$, $\delta = -1$.

In equation (3.21) if one writes $\xi = (y^2 - 1)/(2iy)$, then one gets

$$y'' = \frac{y'^2}{y} - \frac{y'}{\tau} + \frac{1}{\tau}(y^2 + 1) + y^3 - \frac{1}{y}, \tag{3.38}$$

a P3 with $\alpha = 1$, $\beta = 1$, $\gamma = 1$, $\delta = -1$. Expressing y in terms of ξ,

$$y_\pm = i\xi \pm \sqrt{1 + (1\xi)^2}, \tag{3.39}$$

one gets equation (3.32) or (3.33).

Some one-parameter families of solutions of these equations are known, but none of them satisfies our conditions at $\tau = 0$.

Reference

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