Crossover in Low-Temperature Collective Spin-Density-Wave Transport

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We have measured the nonlinear transport in the spin-density-wave (SDW) state of $(TMTSF)_2PF_6$. Besides the typical sliding SDW conduction, at temperatures close to T_{SDW}, we find a new type of collective electron transport below $T \simeq 1$ K, with temperature-independent conductivity which follows a Zener-type expression. We discuss the relevant energy scales of the ground-state excitations, both for single-particle tunneling and for coherent SDW processes.

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Recent experiments performed on various linear-chain organic materials with a spin-density-wave (SDW) ground state [1] establish the essential features of the collective-mode dynamics of this broken-symmetry ground state. Although early experiments on nonlinear transport [2] were controversial, recent measurements [3-6] on high-quality specimens have clearly demonstrated that there is a well-defined threshold field E_T for the onset of nonlinear response; a feature characteristic for collective-mode transport. The threshold field is also usually regarded as evidence that the dynamics can be described on terms of classical variables [7].

Current oscillations [4], with frequency proportional to the excess current, give direct evidence that the current is carried by the translational motion of the condensate. The small threshold field is also accompanied with a large static dielectric constant [5], and with low-frequency or long-time relaxation phenonema. Frequency-dependent conductivity measurements [8] conducted over a broad spectral range also identify both the single-particle excitations across the gap and the collective-mode contribution well below the gap. The above observations are remarkably similar to findings made on charge-density-wave (CDW) condensates [9], in the temperature region where screening by normal carriers are important. In case of charge-density waves at low temperatures, where normal electrons are removed, Coulomb effects lead to a more coherent collective-mode response [10]. This is characterized with sharp nonlinearities and the dynamics of internal degrees of freedom is strongly suppressed [11].

In this Letter we report our transport measurements conducted in wide temperature and electric-field ranges in (tetramethyl-tetraseleno-fulvalene)$_2$PF$_6$, $(TMTSF)_2$-PF_6. The material undergoes a metal-insulator transition at $T_{SDW} = 11.5$ K, and various magnetic measurements clearly suggest a spin-density-wave ground state which is incommensurate with the lattice. We find that at temperatures not far below T_{SDW} the nonlinear response is determined by internal SDW deformations, as in the case of the conventional sliding CDW conduction, but at low temperatures our results are highly suggestive for a new type of collective-mode excitation. Our experiments point to important differences between charge-density-wave and spin-density-wave dynamics, and we discuss the underlying reason [12] for this difference.

Single-crystal specimens used in this study were grown by the standard electrochemical crystal-growth method. Special care has been taken to avoid temperature and current fluctuations during the growth process.

The electrical transport measurements were conducted by a variety of experimental techniques designed to avoid self-heating, artificial contact, or nonuniform current distribution effects. All resistivity curves and current-voltage characteristics were taken in four-probe configuration. Crystals with typical dimensions of $4 \times 0.1 \times 0.1$ mm^3 were investigated. The electrical leads, 10-μm-thick annealed gold wires, were glued on evaporated silver contacts. The current contacts covered the whole area at the crystal ends, and the voltage contacts also surrounded the sample. At low electric fields we used a high-resolution dc method, or lock-in technique with small-amplitude ac modulation. The pulse technique was applied in the high-field experiments; either the field or the temperature was varied. The dielectric response was studied both by ac methods and by real-time relaxation measurements. In the overlapping field, temperature, and frequency ranges the different techniques gave identical results.

In Fig. 1 we show the dc resistivity measured as the function of temperature at various applied electric fields.

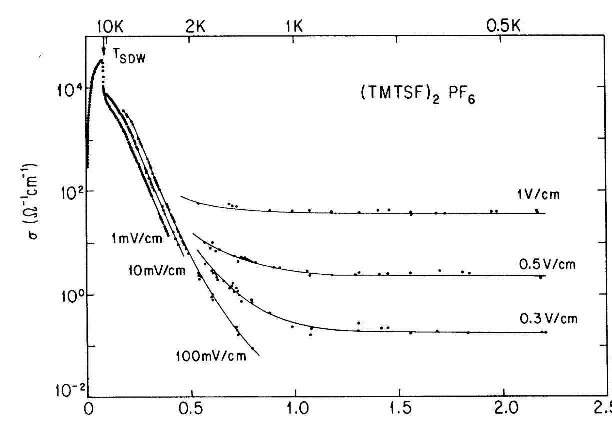

FIG. 1. Temperature dependence of the dc conductivity in $(TMTSF)_2PF_6$ at various electric fields.

The phase transition at $T_{SDW}=11.5$ K is indicated by the arrow. For the smallest applied field the conductivity is Ohmic and at temperatures not much below T_{SDW} it decreases in a fashion characteristic of a semiconductor. In this temperature range the low-field conductivity is determined by single-particle excitations,

$$\sigma_{dc}(T)=\sigma_0 \exp(-\Delta/kT)\,, \tag{1}$$

with $\Delta=23$ K, which is close to the BCS value of $\Delta=1.76kT_{SDW}$, and corresponds to the single-particle gap, also observed in the optical studies [6].

In the temperature range of 1 to 10 K the spin-density wave is depinned [3–6] at electric fields as low as a few mV/cm. Figure 1 shows that above the depinning threshold the contribution of the moving condensate increases the conductivity, but with decreasing temperature the sliding SDW transport freezes out in the same way as the single-particle current. In contrast, at low temperatures the behavior of $\sigma(E,T)$ is fundamentally different. Below 1 K the conductivity is strongly dependent on the applied electric field and is independent of temperature over a broad temperature interval.

A more detailed analysis of the current-voltage characteristics, revealing also the crossover between the two temperature ranges, is given in Fig. 2. The current is Ohmic at low applied fields, but an extra current appears above the sharp threshold electric field E_T, which was found to be only weakly temperature dependent in the range of our experiments. With decreasing temperature both the single-particle, Ohmic current and the sliding component freeze out, and from 5 to 2 K the current drops by several orders of magnitude. Below about 2 K the sliding component decreases considerably faster than the Ohmic component and at $T=1.5$ K no SDW depinning can be detected around the electric-field region where E_T was found at higher temperature. The behavior in this temperature range is in full agreement with recent low-field data of Kriza *et al.* [6].

Figure 2 also demonstrates that at lower temperatures a new type of nonlinear conduction emerges. It is progressively more and more visible as the conventional sliding SDW conduction disappears. In the temperature range of 1.5 to 2.1 K the crossover between the two nonlinear processes is clearly visible with the conventional nonlinear response taken over by a sharp rise of the conductivity at high electric fields of the order of 0.5 V/cm. At even lower temperatures, where the first process is already frozen out, the new nonlinear term overcomes gradually the Ohmic conduction of the normal carriers. The low-temperature limit is characterized with a universal, temperature-independent curve. This feature is also evident from Fig. 1.

The above experiments on $\sigma(E,T)$ provide clear evidence for the presence of two distinct processes: (i) a strongly temperature-dependent SDW conduction appearing above a well-defined threshold field, and (ii) a temperature-independent contribution to the current with a current-voltage characteristic distinctively different from that observed at high temperatures [3–6]. We have also found that, while the high-temperature nonlinear response is sample dependent (as is determined by the residual impurity concentration [Ref. [3(b)]]), the low-temperature response does not show variation from specimen to specimen. This suggests that the nonlinear response is determined by different interactions which involve the SDW ground state. Figure 3 displays the current density as a function of the inverse electric field as measured at $T=435$ mK. The experimental observa-

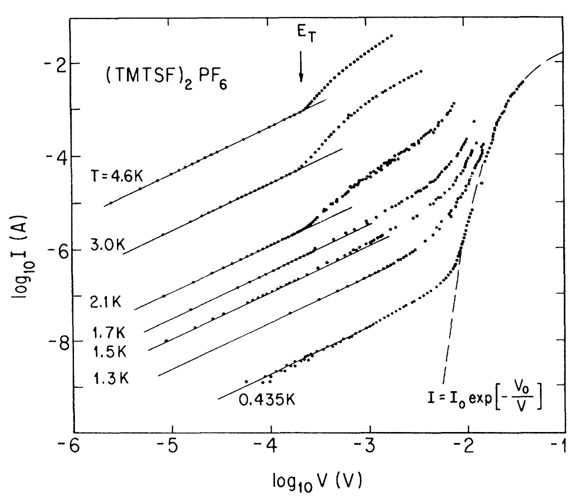

FIG. 2. I-V characteristics of $(TMTSF)_2PF_6$ measured at different temperatures. The solid lines represent the linear relation of normal carriers. The depinning threshold of the first nonlinear process is indicated by the arrow. The new type of nonlinearity, recovered at low temperatures, is described by Eq. (2) (dashed line).

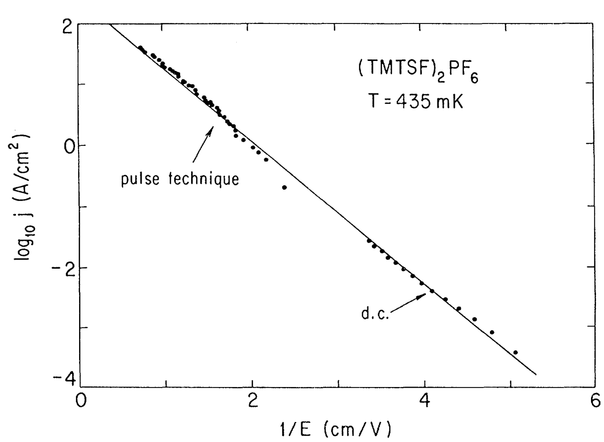

FIG. 3. Nonlinear current vs the inverse applied electric field at $T=435$ mK. The solid line corresponds to Eqs. (2) and (4) with parameters given in the text.

tion can be well described with the Zener formula:

$$j(E) \propto \exp(-E_0/E) . \quad (2)$$

The solid line in Fig. 3 is Eq. (2) with $E_0 = 3$ V/cm. Experiments on three different specimens, drawn from different growths, gave identical results within the error of the crystal size determination (20%).

In the temperature range, where the crossover between the two qualitatively different nonlinearities occurs, there is a significant change in the low-frequency dielectric response as well. Figure 4 displays the temperature variation of the dielectric constant recorded at $\omega/2\pi = 10$ kHz. We have shown recently that the large value of ε reflects the internal SDW deformations and is accompanied with relaxation dynamics characteristic of glassy systems [5]. This response, similarly to the conventional nonlinear SDW conduction, also freezes out below $T \simeq 2$ K, as shown in the figure.

Discussing the results first we note that at temperatures above $T \simeq 2$ K the sliding SDW conduction in $(TMTSF)_2PF_6$ is rather similar to the behavior observed for charge-density waves at temperatures not very far below the phase transition. In this range the extra current scales with the density of thermally excited carriers [13], a feature shown in Figs. 1 and 2. This indicates that screening of the internal deformations by the uncondensed electrons is important and the damping of the collective mode is determined by dissipative normal currents [14]. Moreover, the sharp threshold field found for the SDW conduction in this temperature range is related to the static dielectric constant by the same law as in the case of CDW systems [5]:

$$\varepsilon(\omega \to 0) E_T = c2en , \quad (3)$$

where n is the number of conducting chains per unit area and c is a numerical constant in the order of unity.

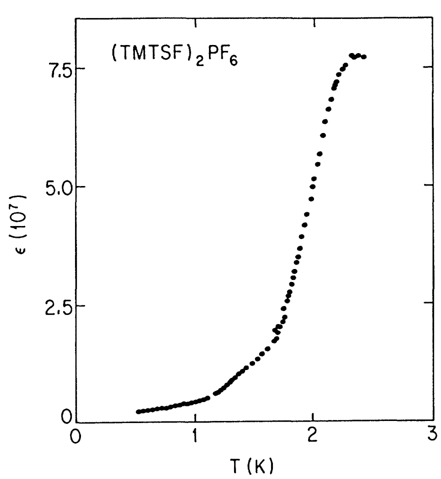

FIG. 4. Temperature dependence of the dielectric constant measured at $f = 10$ kHz.

Equation (3) expresses a general relation between the static polarizability and the depinning threshold and connects the field- and frequency-dependent excitations of the collective mode. In CDW systems it holds at low temperatures, as well, where an abrupt onset of CDW conduction and a collective-mode resonance below the single-particle gap are observed [10]. In this case the sharp threshold field of the low-temperature CDW conduction is related to the dielectric enhancement due to the pinned-mode resonance [10,11]. These two features of the CDW ground state, namely, the steep current rise and the narrow resonance below the gap frequency, have been explained as arising from unscreened CDW mode [15].

In case of spin-density waves the situation is fundamentally different. There are significant differences in the low-temperature nonlinear conduction phenomena. Moreover, there are evidences which show that in the SDW ground state no excitations are present below the single-particle gap; the spectral weight of the resonance found in the GHz frequency range is suppressed at low temperatures as shown by both microwave [8,16] and dielectric [5] studies. It has been suggested [12] that this suppression is due to the Anderson-Higgs mechanism. The freezing out of the low-frequency dynamics with decreasing temperature is also clearly demonstrated by Fig. 4—as according to the Kramers-Kronig relation the dielectric constant $\varepsilon(\omega)$ integrates the contribution of all the higher-frequency modes. The absence of the pinned-mode resonance in the frequency-dependent response indicates that ideas suggested for the low-temperature CDW depinning [15] cannot be fully adopted for the spin-density waves.

It is to be noted that the above experimental results exclude not only the collective-mode resonance but also any significant single-particle excitation below the gap. Low-temperature nonlinearity in principle might arise from tunneling between impurity levels, however, impurity states should also show up both in the low-temperature dielectric constant and in the frequency-dependent conduction over the spectral range below the single-particle gap.

The simplest way to explain the low-temperature nonlinearity would be the assumption of Zener tunneling of individual electrons, a process which emerges as the sliding SDW conduction disappears. Although it would describe the behavior given by Eq. (2), a straightforward numerical analysis makes this possibility unlikely. The characteristic electric field E_0 can be used to evaluate the length scale for the tunneling process, and the WKB approach leads to

$$E_0 = \tfrac{1}{4}\pi^2\Delta^2/E_F e\zeta . \quad (4)$$

E_0 and Δ, derived from Figs. 1 and 3, together with a Fermi energy of the order of $E_F = 1$ eV gives $\zeta = 1100$ Å. This value far exceeds the lattice constant $a \simeq 3$ Å, expected for simple Zener tunneling. Consequently, the nonlinear conduction observed in this field range together with a characteristic distance in order of the SDW coherence length is highly suggestive of collective effects. Equations (2) and (4) also imply that tunneling involving coherent SDW segments cannot be *a priori* excluded.

Macroscopic quantum tunneling has been suggested before for CDW's [17], but the experimental results remain controversial. Because of the small effective mass, tunneling, in principle, is more likely for spin-density waves, but further experiments are required to test this hypothesis. In particular, experiments on alloys could establish whether the characteristic field, E_0, is determined by intrinsic parameters (such as the BCS coherence length) or by impurity-controlled quantities, such as the phase-phase coherence length. Experiments conducted at various pressures, which lead to changes in T_{SDW} and consequently in Δ, could also further test the validity of Eq. (4).

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