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Memory effects in the spin-density-wave state of $(TMTSF)_2PF_6$ (where TMTSF is tetramethyltetraselenafulvalene)

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A memory effect is found in the transient electrical response of the spin-density-wave (SDW) system $(TMTSF)_2PF_6$ (where TMTSF is tetramethyltetraselenafulvalene): the voltage response to a current pulse depends on the electrical history of the sample. The memory persists for times far longer than the characteristic relaxation times of both the single-particle and the collective SDW excitations. We suggest that the memory effect is a manifestation of macroscopically asymmetric metastable pinned spin-density-wave states similar to those observed in charge-density-wave systems.

It is now well established that collective excitation of the spin-density waves (SDW) developing at low temperatures in certain quasi-one-dimensional organic conductors[1] such as $(tetramethyltetraselenafulvalene)_2PF_6$ [or $(TMTSF)_2PF_6$] gives rise to phenomena similar to those found in charge-density-wave (CDW) compounds.[2] In equilibrium the SDW is pinned to the underlying lattice by random impurities. Electric fields as low as a few mV/cm depin the condensate and the sliding mode gives an extra current in addition to that carried by normal carriers.[3] A study of doped and irradiated crystals[4] confirmed the concept of impurity pinning, and the observation of narrow-band noise verified the hypothesis of collective conduction.[5]

Besides the apparent similarities there are also significant differences between the CDW and SDW excitations. The most important is the strongly reduced spectral weight[6] or even the absence[7] of the pinned-mode resonance in the case of spin-density waves. This has been attributed to the Anderson-Higgs mechanism, which is relevant because of the small SDW effective mass.[8] Qualitative differences have been found in the ground-state nonlinear conduction as well.[9]

The randomness and frustration introduced in the system by the interaction of SDW's with randomly positioned impurities and by the large phase-coherence length[10] of the order parameter are expected to give rise to a broad distribution of dielectric relaxation times as well as to metastable pinned SDW states frozen on reasonable experimental time scales. Indeed, a stretched-exponential dielectric relaxation has been observed[11] recently in the SDW state of $(TMTSF)_2PF_6$. In CDW sytems, the metastable pinned density-wave states are observed experimentally via memory effects, i.e., through the dependence of various physical properties on the electric or thermal history of the sample. Perhaps the most spectacular of these phenomena is the so-called pulse-sign memory effect: the voltage response of the system to a current pulse depends on the polarity of the previously applied pulse. This effect was first observed by Gill[12] in $NbSe_3$ and later it was found in most CDW systems.[13,14] It was also shown that temporary depinning of the CDW leads to macroscopic asymmetry in the specimen and various electric properties may reflect the direction of the previously applied field.[15,16]

In this paper we present the observation of the pulse-sign memory effect in a spin-density-wave system, $(TMTSF)_2PF_6$. Our results demonstrate that the pinned state of the SDW condensate is not uniquely defined and indicate the existence of electric-field-induced macroscopic asymmetry in pinned spin-density waves.

Single crystals of $(TMTSF)_2PF_6$ were prepared by standard electrochemical crystal growth methods. Crystals drawn from different batches showed the same phase transition temperature of $T_{SDW} = 11.5\,K$, while the threshold field for SDW depinning was in the range of $E_T = 2$ to $4\,mV/cm$ at $T = 4.2\,K$. The electric memory effects have been found in all specimen investigated.

In Fig. 1 we demonstrate the pulse-sign memory phenomenon in the spin-density-wave state by showing voltage responses to double current pulses of opposite and alike polarities. The first current pulse results in an electric field of magnitude slightly above E_T, while for the second the field is below the threshold. The dashed line shows that the voltage response to the second pulse is fast if the preceding pulse has the same polarity. If, however, the preceding pulse has an opposite sign, the response is sluggish and the voltage rise extends over several milliseconds. This is shown by the continuous line. The steady-state value corresponds to the same normal conductivity as before, but the difference between the two curves signifies different initial states. Note that at the trailing edge the two responses are identical.

The difference in the transient responses has been interpreted[17] in CDW systems as arising from a macroscopic charge displacement within the sample upon the reversal of the polarity of the field. Then the integrated area between the two traces corresponds to the charge

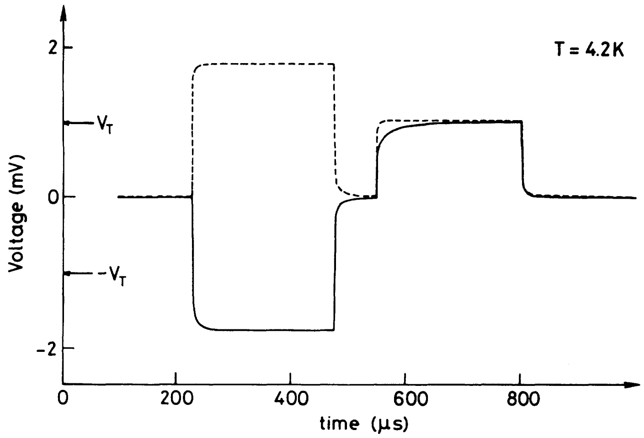

FIG. 1. Voltage response to current pulses in the spin-density-wave state of $(TMTSF)_2PF_6$. The dashed line is for unipolar pulses. The full line is the voltage trace if the second pulse has an opposite sign than the first pulse. The transient at the second pulse shows a slow discharging process if previously the SDW was depinned in the opposite direction. The arrow shows the threshold for the nonlinear conduction.

involved in the process and characterizes the magnitude of the memory effect. We refer to this integral as a pulse-sign memory (PSM) signal. In the experiment shown in Fig. 1, the PSM signal is in the order of one electron per conducting chain, a value comparable to that observed in charge-density-wave compounds. An accurate determination of the displaced charge is difficult for at least two reasons. First, the line shape is nonexponential and the contribution of the hardly resolved long-time tail to the integral may be important, and second, spatial inhomogeneities within the sample may distort the results. For the sake of consistency, in the following we use charge values obtained from integrals performed over the time interval of 10^{-5} to 10^{-2} s.

The influence of the previous pulse extends to extremely long times. Figure 2 demonstrates that the pulse-sign memory effect is well observable even if the waiting time between pulses of opposite sign is as long as 100 s. The data shown in the figure were collected at $T = 2$ K, where the dielectric behavior of the pinned spin-density-waves has also been explored in detail.[11] For comparison, the time constant of the single-particle excitations at this temperature is $\tau_{sp} \sim 10^{-8}$ s, while the mean relaxation time for the collective mode response is $\tau_{SDW} \sim 10^{-3}$ s. Therefore in zero electric field the pulse-sign memory is preserved for times at least five orders of magnitude longer then the characteristic times of dielectric relaxation. On the other hand, in an electric field close to E_T the transition from one state to the other—as observed via the sluggish response upon sign reversal—occurs on time scales comparable to τ_{SDW}, and may well be related to the stretched-exponential relaxation found in the system.

The magnitude of the effect depends on the applied electric field. Although it is best visible in the vicinity of the threshold field, crossing E_T is not necessary for the observation of the pulse-sign memory. In Fig. 3 the integrated excess current is plotted as a function of electric field. In this experiment we applied symmetric pulses, i.e., only the sign of the pulses varied during a pulse sequence at a fixed field value. No singularity is found at E_T in accordance with results obtained in charge-density-wave systems.[14] We do not attribute any significance to the broad peak seen above the threshold, either. Since the relaxation may be accelerated by application of electric fields, as in case of CDW's,[18] the high-field part of the curve might also be suppressed as a calculation artifact due to the fixed limits in the integral. The general behavior shown on the figure is rather similar to that found in TaS_3 (Ref. 14).

In order to explain the electric memory effects in charge-density waves it has been assumed[17] that under the influence of an external field the condensate deforms, and this deformation is preserved in the pinned state, even after the field is turned off. In the case of unipolar pulses the distortion—frozen in the pinned state—is always the same. However, in reversal of the field a transition occurs from one distorted state to another, accompanied by macroscopic charge displacement within the sample. The extra displacement current gives rise to the unusual transient, which appears only when the field direction is reversed. Since our experiments on $(TMTSF)_2PF_6$

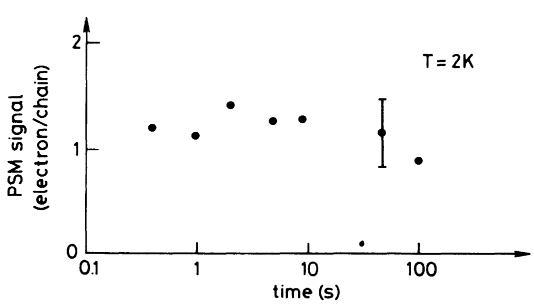

FIG. 2. The accumulated charge associated with the memory effect as a function of the waiting time between two pulses. The pulse width was 1 ms.

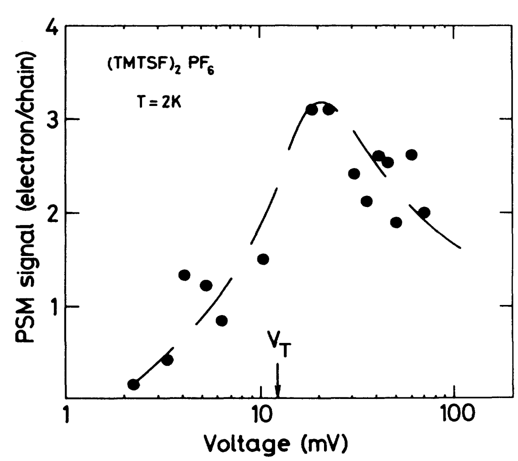

FIG. 3. The accumulated charge as a function of applied field. The threshold is shown by arrow. The dashed line is to guide the eyes.

were carried out in the field and temperature range where the concept of random pinning of incommensurate condensate is valid, we believe that the above picture can be adopted to spin-density waves as well.[19]

We note that the assumption[17] of field-induced macroscopic asymmetry has been experimentally confirmed for CDW's. It was shown that in the pinned state the order parameter is nonuniform, and its spatial variation depends on the direction (and magnitude) of the previously applied field.[15,16] Moreover, thermally stimulated discharging measurements have directly indicated a finite polarization attributed to distortions of pinned CDW's.[20]

In conclusion we have shown that the transient response of $(TMTSF)_2PF_6$ depends on the electric history of the sample. The memory effect found in the spin-density-wave salt is phenomenologically identical to that observed in charge-density-wave compounds. Our results indicate macroscopic deformations of spin-density waves.

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