Dielectric Relaxation of the Pinned Spin-Density Wave in $(TMTSF)_2PF_6$

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The low-frequency dielectric response has been measured in the spin-density-wave state of $(\text{tetramethyltetraselenafulvalene})_2PF_6$. The dielectric constant is significantly larger than that expected for single-particle excitations and is dominated by the internal modes of the spin-density-wave condensate. We also establish the relation between the frequency-dependent and nonlinear response.

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Several aspects of the dynamics of the broken ground state, called the spin-density wave[1] (SDW), have been explored recently in the Bechgaard salts, of which the material $(\text{tetramethyltetraselenafulvalene})_2PF_6$ $[(TMTSF)_2PF_6]$ is the most prominent example. The high-frequency electrodynamics is characterized[2] by single-particle excitations across the gap Δ and by a low-lying mode, which appears at a frequency around 5 GHz. The latter can be associated with the resonance of the pinned condensate. The spectral weight of this collective mode is unexpectedly small indicating either a large dynamic mass or a small number of relevant excitations.[3] While early conductivity measurements have shown that the dc transport is nonlinear,[4] a sharp threshold field for nonlinear conduction has been found only recently in high-quality specimens.[5-7] Another signature of collective-mode transport, current oscillations (alternatively known as narrow-band noise), have also been detected in the SDW state of some Bechgaard salts.[7,8]

In this paper we report our experiments on the low-frequency dielectric response in the SDW state of $(TMTSF)_2PF_6$. We find direct evidence for a collective-mode contribution to the dielectric properties and establish the relation between the static polarizability and the depinning threshold field. Furthermore, our real-time relaxation experiments are the first to reveal internal spin-density-wave excitations. The observed glassy behavior indicates a wide distribution of metastable pinned SDW configurations. Our data are also suggestive of a small spectral weight for the pinned mode resonance, which occurs above the frequency domain of this study.

High-purity $(TMTSF)_2PF_6$ single crystals originating from different sources were measured. One batch was prepared at the Université Paris-Sud, Orsay, and was supplied to us by D. Jérome and P. Batail, and one originated from F. Wudl's group at the University of California, Santa Barbara. Both batches displayed rather similar behavior, as will be discussed below. The typical size of crystals used in this study was $3\times0.05\times0.03$ mm^3.

Electrical contracts were made by mechanically clamping 0.01-in. wires to gold pads evaporated on the crystal.[6,9] We employed a slow cooling rate of about 0.1 K/min. We did not find any sudden jumps in the resistance, indicating that no breaks were generated during the cooldown. The measurements were reproduced in a two-probe configuration on a sample where the crystal ends were also covered by gold, ensuring homogeneous current injection. The experiments presented below were performed at $T=2$ K, where the sample resistance dominated over the contact resistances. The four samples studied have a conductivity of $\sigma(2\text{ K})\simeq 10\ \Omega^{-1}\text{cm}^{-1}$, within a factor of 2, in accordance with previous results.[10]

In Fig. 1 we show the nonlinear conductivity measured at $T=2$ K. At low electric fields (where heating effects

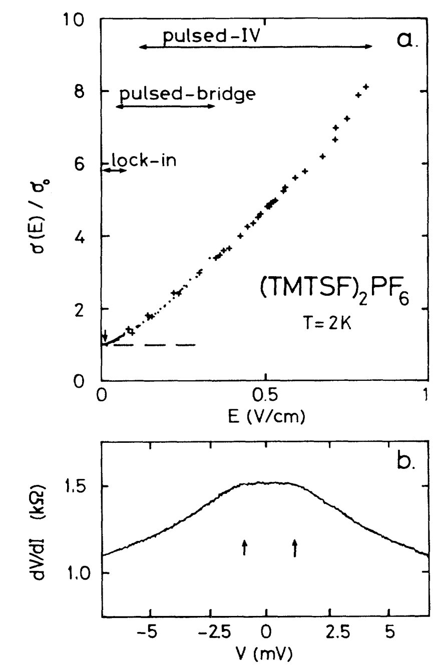

FIG. 1. SDW depinning by electric field in $(TMTSF)_2PF_6$ at $T=2$ K. (a) Field dependence of the conductivity measured by various methods. (b) Differential resistance in the vicinity of the threshold voltage. (The arrows indicate the depinning threshold in both parts.)

are negligible) the differential resistivity was detected with a lock-in. A pulsed bridge configuration was applied in intermediate fields, while for large voltage amplitudes, short pulses (10–100 μs) were used and the resulting current was measured. Similarly to charge-density waves,[11] and earlier studies[6-8] on various $(TMTSF)_2X$ salts, we find a sharp threshold field. The specimen on which nonlinear conduction is shown was prepared in Orsay and has a threshold field of $E_T = 12$ mV/cm. In the batch prepared at Santa Barbara we also obtained $E_T \simeq 10$ mV/cm, but the nonlinearity was somewhat less pronounced.

A variety of experimental techniques have been employed to explore the frequency- and field-dependent response. In a wide frequency range $\varepsilon(\omega)$ was obtained by Laplace transformation from real-time experiments:

$$\varepsilon(\omega) = 4\pi \int p'(t)\cos(\omega t)\,dt\,. \qquad (1)$$

Here $p'(t)$ is the time derivative of the polarizability, i.e., the charging current density following an electric-field step of unity. To ensure the applicability of linear response theory, care was taken that the current density did not exceed the threshold value.

In the 10-kHz to 0.5-MHz range, a conventional lock-in technique was applied, and experiments with various driving amplitudes established that the ac response represents the low-field limit. Data presented below were recorded at μV levels, i.e., below 1% of the depinning threshold. For the experiments in the GHz range, the sample was mounted on the end of a 50-Ω waveguide and the impedance was monitored as a function of the frequency in a compensated bridge circuit using an HP-8754A network analyzer.

Figure 2 shows the time dependence of the discharge current $j(t)$ detected when the electric field E was switched to zero. [Note that $j(t)/E = p'(t)$ in Eq. (1).] The solid line in the figure corresponds to an empirical expression for p:

$$p(t) = p_0 \exp[-(t/\tau)^{\alpha}]\,, \qquad (2)$$

the so-called stretched exponential form, with $\tau = 1.1$ ms and $\alpha = 0.67$. Such an expression has been extensively used to describe relaxation processes in various glasses and random systems.[12,13] The experimentally found nonexponential relaxation (generally associated with disorder) reflects a broad distribution of the relaxation times.

In Fig. 3 we display the dielectric constant evaluated by using the experimental techniques discussed before. The magnitude of ε measured with a lock-in in the 10–100-kHz range agrees within 30% with the value determined from the real-time relaxation data. Note that experiments conducted on specimens from different preparations (denoted as "Orsay" or "Santa Barbara" in the figure) give similar results in both the magnitude and frequency dependence of ε. In the low-frequency data, obtained by Laplace transformation, a systematic error may arise from the finite current resolution of the long-time relaxation measurements. This is indicated by error bars in the figure. The solid line corresponds to the Laplace transform of the stretched exponential expression Eq. (2).

First we discuss the magnitude of the dielectric constant. In a simple semiconductor, the single-particle contribution to the dielectric constant can be expressed in terms of the plasma frequency and the band gap. In $(TMTSF)_2PF_6$ both quantities have been measured.[2]

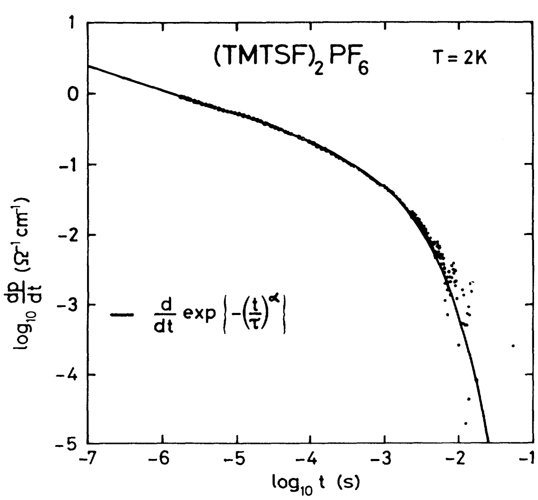

FIG. 2. Time dependence of the discharge current (i.e., the derivative of the polarizability) following a voltage step. The solid line corresponds to a stretched exponential behavior, Eq. (2) with $\tau = 1.1$ ms and $\alpha = 0.67$.

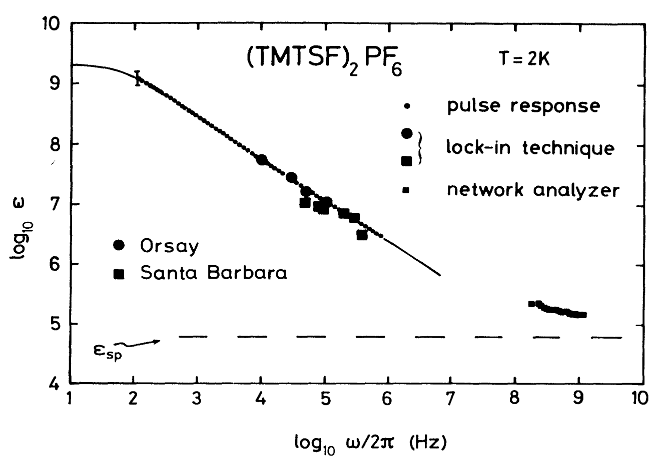

FIG. 3. Low-frequency dielectric constant in the SDW state of $(TMTSF)_2PF_6$. The solid line is obtained by Laplace transformation from the real-time relaxation data shown in Fig. 2. The dashed line indicates the single-particle contribution to the dielectric response.

With $\Omega_p = 10^4$ cm^{-1} and $\Delta = 15$ cm^{-1} one obtains

$$\varepsilon_{\text{s.p.}} = \varepsilon(\omega \ll \Delta/h) = \tfrac{1}{6}(\Omega_p/\Delta)^2 = 6\times10^4 . \quad (3)$$

Another contribution comes to the static dielectric constant from the phase-coherent resonance[2] of the SDW observed around $\omega_0/2\pi \cong 5$ GHz. This mode gives rise to

$$\varepsilon_p = \varepsilon(\omega \ll \omega_0) = S/\omega_0^2 , \quad (4)$$

where $S = \int \sigma(\omega) d\omega$ is the spectral weight of the mode. In a simple harmonic oscillator model the oscillator strength is $S_0 = 4\pi n_{\text{SDW}} e^2/m_b = \Omega_p^2$, where n_{SDW} is the number of electrons condensed into the collective mode and m_b is the band mass [as no mass enhancement is expected for SDW (Refs. 15 and 16)]. This dielectric contribution should not depend on ω in the range of our experiments. Figure 3 indicates that such a frequency-independent contribution must be smaller than 2×10^5, the value measured around 1 GHz. Consequently, Eq. (4) suggests a strongly reduced spectral weight of about $S/S_0 \simeq 10^{-4}$. The reduction of the spectral weight has also been suggested earlier from the analysis of micrometer and millimeter wave experiments (performed at a somewhat different temperature).[2] The observation of a small spectral weight is in clear contrast to what is expected for a spin-density wave,[15,16] and may be due to a —yet unexpected—large effective mass or due to Coulomb effects.[3]

The observed low-frequency dielectric constant far exceeds the contribution from the single-particle excitation and indicates a large polarizability for the spin-density waves. The static value of the dielectric constant, obtained by fitting our experimental results with Eq. (2),

$$\varepsilon_0 = \varepsilon(\omega \to 0) = \left.\frac{dp}{dE}\right|_{E\to 0} = 2\times10^9 , \quad (5)$$

can be directly related to the threshold electric field, and various models developed for charge-density waves lead to[16]

$$\varepsilon_0 E_T = c2en_\perp , \quad (6)$$

where $n_\perp$ is the number of conducting chains per unit area and c is a numerical constant of the order of unity. Figure 4 shows that this relation is valid for a spin-density waves as well, confirming the concept of impurity pinning of the collective model.

Finally, we discuss the most striking observation in Figs. 2 and 3, the fact that the dielectric response indicates a broad distribution of modes spread over several decades. We believe that the deviation from a simple exponential decay reflects disorder effects and suggests that spin-density waves are pinned by randomly positioned impurities, with a distribution of relaxation times due to a wide scale of metastable pinned configurations. It is to be noted that the exponent $\alpha = \frac{2}{3}$ of the stretched exponential we observed is close to that found in case of charge-density waves.[13]

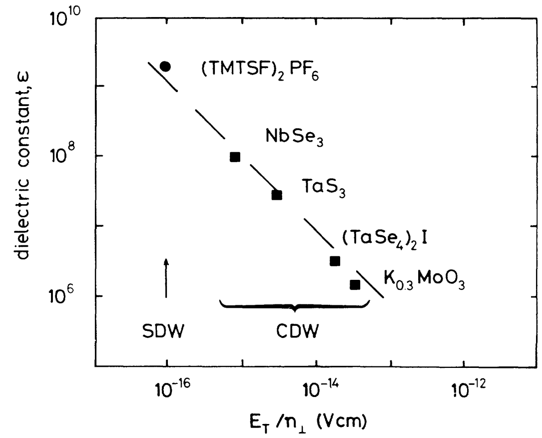

FIG. 4. The relation between the static dielectric constant and the threshold field in $(TMTSF)_2PF_6$ and in various CDW materials (taken from Ref. 14). The dashed line corresponds to Eq. (6) with $c = 0.5$.

In conclusion, we have explored the low-frequency dielectric response of pinned SDW condensate. There are striking similarities to CDW response, but our experiments are also suggestive for a reduced spectral weight of the pinned mode resonance. We have found evidence for low-lying excitations of the internal degrees of freedom in a spin-density-wave system.

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