Shapiro Interference in a Spin-Density-Wave System

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We report on the observation of interference between intrinsic voltage oscillations generated by sliding spin-density waves and an externally applied ac current in $(TMTSF)_2AsF_6$ (TMTSF denotes tetramethyltetraselenafulvalene).

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The understanding of the collective response[1] of spin-density waves (SDW's) to electromagnetic excitations has progressed significantly in the last few years. While the SDW state occurs in a variety of materials, the most prominent model systems are in the family of organic linear-chain compounds (tetramethyltetraselenafulvalene)$_2X$ or $(TMTSF)_2X$ (Ref. 2). The systems with certain anions ($X = NO_3, PF_6, AsF_6, SbF_6$) undergo a metal-insulator transition in which the whole Fermi surface is destroyed by the formation of SDW's.

The first piece of evidence[3] of a collective SDW response was the observation of frequency-dependent conductivity in $(TMTSF)_2PF_6$ at frequencies much smaller than the gap in the single-particle excitation spectrum. Not much later Tomic, Cooper, and Jérome[4] observed a nonlinear dc conductivity above a well-defined threshold field in $(TMTSF)_2NO_3$. The threshold field has been shown[5] to be proportional to the concentration of defects created by x-ray irradiation, proving the importance of lattice defects in SDW pinning.

Even more convincing evidence of a collective SDW response may arise from the observation of coherent effects like voltage oscillations in the nonlinear region or interference phenomena in the presence of combined ac and dc excitations. These effects are general and distinctive features of charge-density-wave (CDW) conduction,[6] and—by the close analogy between CDW's and SDW's—their presence is also expected in SDW systems.

The voltage response of a CDW system to a dc current above the nonlinear threshold contains[7] an oscillating component, often referred to as narrow-band noise, with a frequency ν_0 proportional to the current density j_{CDW} carried by the CDW. Assuming that during one period of the oscillations the CDW (or SDW) is displaced by one wavelength, the proportionality constant is given in the low-temperature limit by

$$j_{\mathrm{CDW(SDW)}}/\nu_0 = 2en_\perp\,, \tag{1}$$

where $n_\perp$ is the number of conducting chains crossing unit cross-sectional area. Equation (1) has been verified[8,9] in CDW systems with high accuracy. Although the origin of these oscillations is still debated, most probably they arise[10] from the creation and motion of topological defects of the CDW condensate. Recent conduction noise measurements by Nomura *et al.*[11] in the SDW state of quenched $(TMTSF)_2ClO_4$ indicate the presence of a peak in the noise spectrum, which moves towards high frequencies upon increasing the current.

If a CDW sample is driven by an alternating current with frequency ν_1 superimposed on a dc bias, an interference is observed[12] between the two frequencies ν_0 and ν_1 whenever their ratio can be expressed as the ratio of two small integers: $\nu_0/\nu_1 = p/q$. The interference appears as a peak in the differential conductivity as a function of electric field. Such a peak corresponds to a step in the current-voltage characteristics, which—by a rather formal analogy with the Shapiro effect in Josephson junctions—is often called a Shapiro step. If the ratio p/q is integer, the interference feature is called "harmonic"; otherwise it is "subharmonic."

In this paper we report on the observation of both harmonic and subharmonic interference in the spin-density-wave conductor $(TMTSF)_2AsF_6$. Based on an estimate of the current distribution in the sample, supported by the measured spectral distribution of the conduction noise, we argue that the ratio j_{SDW}/ν_0 may agree with the expectation described by Eq. (1).

We have studied three $(TMTSF)_2AsF_6$ single crystals prepared by P. Batail and co-workers using the standard electrochemical growth procedure. The crystals have regular rectangular shapes with typical dimensions of 2 mm$\times$100 μm$\times$50 μm. Four electrode contacts have been applied to the samples by first evaporating gold pads on one or four crystal surfaces, and then tightening 12.5-μm-diam gold wires against the pads by soft springs.

No spurious resistance jumps have been observed upon cooling the sample. The SDW transition temperature is $T_c = 11.5$ K with a transition width of 0.2 K, defined as the width at half maximum of the peak in $-d(\ln R)/dT$. The ratio of the room-temperature resistance to the resistance minimum above T_c is about 150.

The nonlinear conductivity below T_c has been investigated either by a pulsed-current method, or—when pulsed measurements indicated that the self-heating of the sample was negligible up to a given current—by a continuous-current lock-in technique. In the latter case

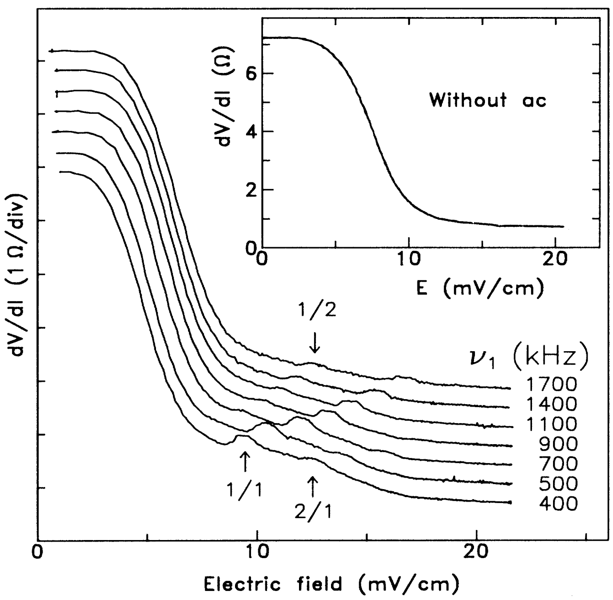

FIG. 1. Differential resistance vs electric field with an applied ac current of frequency ν_1 ($T=4.2$ K). Three different interference features are indicated by arrows. The curves are shifted with respect to each other for clarity. Inset: dV/dI vs electric field without ac current.

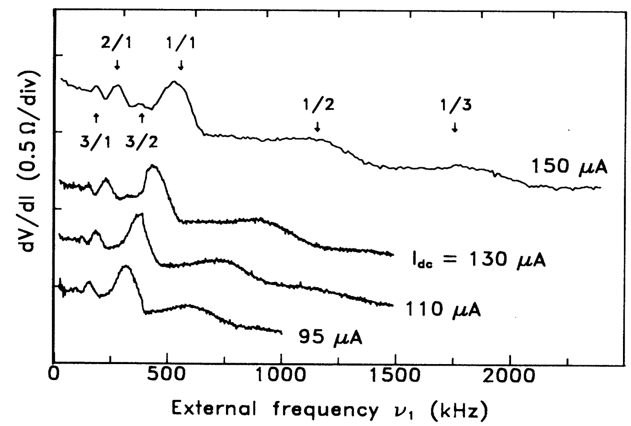

FIG. 2. Differential resistance vs frequency of the applied ac current for constant dc current I_{dc} ($T=4.2$ K).

the current on the sample was modulated at a low frequency (83 Hz), and the differential resistance dV/dI was measured by the lock-in amplifier as a function of the time-averaged current measured on a reference resistance. The sample voltage was then determined by integrating the dV/dI curve. The amplitude of the current modulation was about 1% of the nonlinear threshold.

The electric-field dependence of the differential resistance at 4.2 K is shown in the inset of Fig. 1. The resistance is constant at low fields, and a strong nonlinearity sets in above a fairly sharp threshold field $E_T \approx 5$ mV/cm, a value close to those measured[5,13] on high-purity $(TMTSF)_2PF_6$ samples. The threshold field increases monotonically with temperature. A detailed characterization of the nonlinear conductivity in a wide temperature and electric-field range will be published elsewhere.

To observe the interference between an externally applied frequency ν_1 and the internally generated frequency ν_0 the sample has been driven by a radio-frequency current I_{ac} superimposed on a dc bias I_{dc}, and the differential resistivity has been measured by the lock-in technique described above using a third, low-frequency, low-amplitude current. dV/dI versus electric-field curves measured with different ac frequencies are shown in Fig. 1. The amplitude of the ac current was 220 μA, about 4 times the threshold current. At least three interference peaks are clearly identified in the spectra. The peaks move towards high dc currents as the frequency is increased. No complete mode locking is observed, i.e., the peak value of the differential resistance at the interference is smaller then the low-field "normal" resistance R_n.

Another representation of the phenomenon is obtained when the differential resistivity is measured as a function of ν_1 at a constant dc current. Such frequency scans are shown in Fig. 2. Since the background is flatter in this case, even more interference peaks are resolved. As the SDW current changes only slightly with ν_1, the peaks are easily indexed on this plot. With the reasonable assumption that the largest peak corresponds to $\nu_1/\nu_0 = 1/1$, we identify the peaks $\nu_1/\nu_0 = 1/3$, 1/2, 1/1, 3/2, 2/1, and 3/1.

The nonlinear current carried by the SDW's is calculated as $I_{SDW} = I - V/R_n$. Figure 3 shows the SDW current at the interference peaks indexed as 1/2, 1/1, and 2/1 as a function of ν_1. All three curves are fairly linear

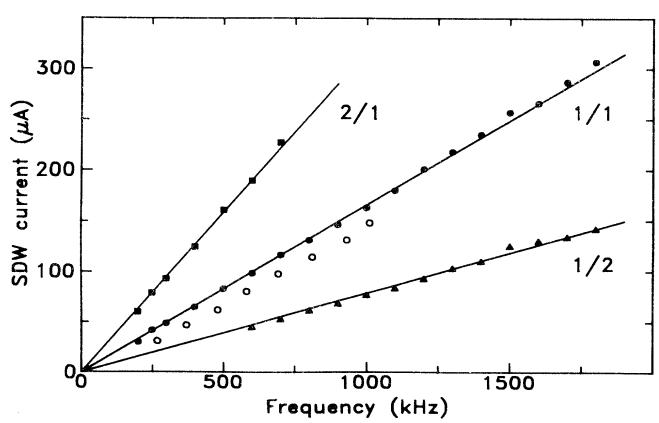

FIG. 3. Solid symbols: SDW current at the interference features $\nu_0/\nu_1 = 1/2$, 1/1, and 2/1 as a function of the frequency of the applied ac current. The ratio of the slopes of the straight lines is 2:1:0.48. Open symbols: SDW current vs the frequency of the peak in the conduction noise spectrum.

and the ratio of the slopes of the linear fits is 2.0:1:0.48.

The internally generated frequency ν_0 that leads to the interference can be observed directly, i.e., without an externally applied ac current by measuring the frequency spectrum of conduction noise. Noise spectra obtained by Fourier transforming time records of the voltage response to various dc currents are shown in Fig. 4. The noise power is distributed over a wide frequency range from zero to an upper cutoff frequency. The noise amplitude increases quasimonotonically with frequency and has a maximum at the cutoff frequency. The position of this peak is indicated together with the position of the interference features in Fig. 3. The noise peak is close in frequency to the 1/1 interference feature. The slight difference is easily attributed to the increased coherence of the conduction noise in the presence of the ac current, a phenomenon well known in CDW systems.[8]

By counting the conducting chains crossing unit cross-sectional area, Eq. (1) yields a current-to-frequency ratio of $j_{\rm SDW}/\nu_0 = 3.1\times10^{-5}$ A cm^{-2}/Hz. Estimating the SDW current density as $j_{\rm SDW} = I_{\rm SDW}/S$, where S is the cross section of the sample, we get $j_{\rm SDW}/\nu_0 = 1.5\times10^{-6}$ A cm^{-2}/Hz, i.e., about 20 times smaller than the prediction of Eq. (1). However, the wide distribution of the conduction noise power and the incompleteness of the mode locking suggest an inhomogeneous current distribution along the sample cross section. In fact, the inhomogeneity is a direct consequence of the high conductivity anisotropy and the contacting method of injecting current on one side of the crystal only.

The current distribution in the linear region is easily calculated by solving the boundary problem in a simple model where the sample is infinite in the **a** direction and semi-infinite in the **c** direction (best and worst conducting directions, respectively), and the current is injected along two lines on the (**a**,**b**) surface parallel to the **b** direction. In the plane halfway between the current injection lines—where the current distribution is the most homogeneous—the current density is maximal at the (**a**,**b**) surface and decays as a Lorentzian as a function of the distance from the surface. The penetration depth of the current defined as the half-width of the Lorentzian is $\lambda = (L/2)(\sigma_c/\sigma_a)^{1/2}$, where L is the distance between current injection lines, and σ_c and σ_a are the conductivities in the respective directions. Taking $\sigma_c/\sigma_a = 10^4$ (Ref. 14) and the actual distance between the current contacts of our sample, $L = 1.6$ mm, the penetration depth is $\lambda = 8$ μm, i.e., about 10% of the thickness of the sample, $t = 75$ μm. This calculation is in agreement with the measured room-temperature (RT) resistance. Assuming a homogeneous current distribution, $\sigma_{\rm RT} = 120$ $(\Omega\,{\rm cm})^{-1}$ is obtained, much smaller than $\sigma_{\rm RT} \approx 700$ $(\Omega\,{\rm cm})^{-1}$ measured by silver-painted contacts covering all sides of the sample.[2] Using the inhomogeneous current distribution calculated in the surface-contact model, however, we recover a conductivity $\sigma_{\rm RT} = 770$ $(\Omega\,{\rm cm})^{-1}$.

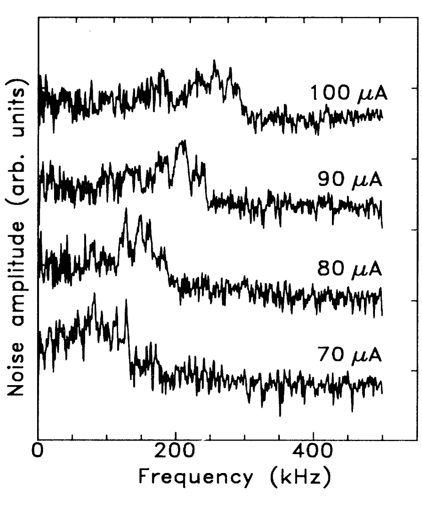

FIG. 4. Conduction noise spectra for various dc currents. A base line measured with zero current is subtracted from the spectra. The threshold current for nonlinear conductivity is $I_T = 50$ μA. ($T = 4.2$ K.)

The distribution of the SDW current is much more difficult to estimate, not only because of the nonlinearity of the problem, but also because of the *a priori* unknown nonlocal dependence of $j_{\rm SDW}$ on electric field. The observed conduction noise spectra suggest an SDW current distribution extending from zero to a maximum, in agreement with the surface-contact model. The presence of the peak at the high-frequency end of the spectrum is also understood since the current distribution is the most homogeneous at the surface where $j_{\rm SDW}$ is the highest. It is also clear that because of the finite threshold field and the nonlinearity of conductivity, the SDW current is even more inhomogeneous than the normal current. The measured $I_{\rm SDW}/\nu_0$ ratio may then well be in agreement with Eq. (1).

A trivial way to improve the homogeneity of the current distribution is to apply the gold pads for current injection to all sides of the crystal instead of only one surface. We have measured two samples prepared this way. In both cases, however, the conduction noise spectra consisted of several peaks not harmonically related. Increasing the current, more and more new peaks appeared at low frequencies even when the electric field was several times higher than the nonlinear threshold, indicating a partial depinning of SDW's even in such high electric fields.

In conclusion, we have measured the interference between the voltage oscillations generated by sliding SDW's and an externally applied ac current. All main features of the phenomenon observed in CDW systems exist in the SDW system investigated. Both harmonic and subharmonic interference features are observed with frequencies proportional to the current carried by the

SDW. The frequency of the fundamental interference is close to the frequency of the peak in the conduction noise spectrum. As the SDW current flowed in only a fraction of the sample cross section, no quantitative determination of the current-density to noise-frequency ratio was possible. An estimate of the current distribution, however, suggests that the ratio may be equal to that observed in CDW systems. The new data presented in this study provide firm evidence for the existence of a novel collective transport in a SDW condensate.

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