Conduction Noise and Motional Narrowing of the Nuclear Magnetic Resonance Line in Sliding Spin-Density Waves

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We report on joint conduction noise and ^{13}C NMR measurements in the sliding spin-density wave (SDW) state of $(\mathrm{TMTSF})_2\mathrm{PF}_6$, where TMTSF is tetramethyltetraselenafulvalene. The noise distribution is narrow and scales linearly with current. Narrowing of the NMR line was observed simultaneously. The comparison of these two sets of data shows that the sliding motion is highly coherent and that the conduction noise frequency is equal to the winding rate of the SDW. In the depinned volume, a substantial fraction of the conduction electrons participate in the sliding mode. Consequences on the origin of conduction noise and on other transport measurements are discussed.

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The comparison of the dynamics of incommensurate spin-density waves (SDW) with charge-density waves (CDW) in quasi-one-dimensional metals has aroused a lot of interest recently. By now, all the main features typical of incommensurate CDW's have been evidenced in SDW's of the molecular conductors $(\mathrm{TMTSF})_2X$ (X is a monovalent anion, e.g., PF_6) [1]. Although the phase ϕ of the SDW order parameter is pinned to crystalline defects, the phase mode dominates the linear dielectric [2] and magnetic [3] response at low frequencies. A spectacular nonlinear conductivity has also been demonstrated above a sharp threshold electric field [4]. This nonlinearity is attributed to the depinning and "sliding" of the SDW.

If the sliding is described as a simple translation of the SDW, the SDW velocity is characterized by the phase winding rate $\nu_\phi \equiv (2\pi)^{-1}\partial\phi/\partial t$, and the current carried on each conducting chain by the sliding density wave (DW) is

$$j_{\mathrm{DW}} = 2ef_c\nu_\phi \,, \tag{1}$$

where f_c is the condensate density, which is expected to tend to 1 at low temperatures.

The sliding of the density wave results in a coherent temporal modulation of the local magnetic (for SDW) or electric (for CDW) fields. Since in the static DW, the NMR line shape is broadened by the spatial inhomogeneity of the local fields, the sliding of the DW averages out the inhomogeneity and induces a narrowing of the NMR line, which provides a measurement of ν_ϕ . This effect has been observed both in CDW's [5] and SDW's [6].

The DW sliding also generates conduction "noise"; in high-quality CDW samples it is, in fact, a coherent voltage oscillation [7] (hence the ambiguous notion narrow band noise) with frequency ν_n proportional to j_{DW}. Despite an intensive research in the last decade, the origin of these oscillations is still controversial [8]. Several models of noise generation suggest $\nu_n \propto \nu_\phi$, but the proportionality constant is model dependent, usually 1 or 2.

Measured j_{DW}/ν_n values for CDW's are in agreement [9] with Eq. (1) and $\nu_n = \nu_\phi$. Moreover, by a simultaneous measurement of NMR and conduction noise, Jánossy *et al.* [10] have been able to prove both $\nu_\phi = \nu_n$ and $f_c = 1$ in the CDW state of $\mathrm{Rb}_{0.3}\mathrm{MoO}_3$.

In SDW's, a conduction noise has also been observed [11], but j_{DW}/ν_n has been found to be 8 to 20 times smaller than expected. This discrepancy may arise from an erroneous estimate of j_{DW} due to a strongly inhomogeneous current distribution as seen from the broad noise spectra. However, it has been also proposed—based on the small oscillator strength [12] found in a high-frequency phase mode, the so-called pinned mode resonance—that the number of excitations relevant for the phase mode is small, or, equivalently, instead of $f_c = 1$, an effective condensate density $f_c \ll 1$ should be taken in Eq. (1).

In this paper we report on measurements of the current-voltage characteristics, conduction noise, and ^{13}C NMR in the SDW phase of $(\mathrm{TMTSF})_2\mathrm{PF}_6$. We obtain a well-defined conduction noise frequency distribution. The mean value increases linearly with the SDW current. From a comparison of noise and NMR spectra taken at the same currents, we conclude that the noise frequency is equal to the phase winding rate. From the NMR measurements we determine the volume in which the SDW is depinned, and find that in this volume all the electrons participate in the collective sliding mode within the experimental uncertainty. We discuss the consequences of these results on the possible mechanisms of narrow band noise generation as well as on other transport properties.

We have investigated the transport and NMR proper-

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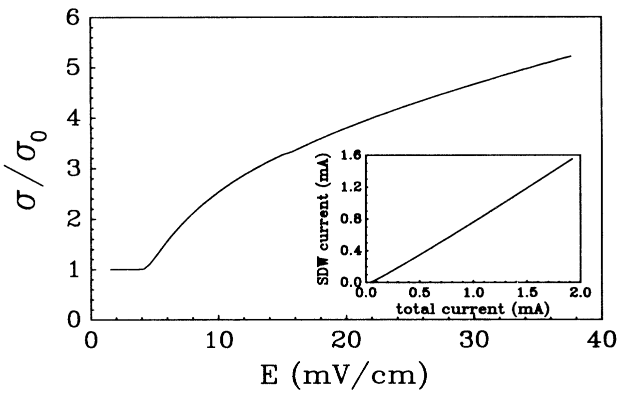

FIG. 1. Conductivity σ as a funtion of electric field E, normalized to the low-field value σ_0. Inset: SDW current as a function of the total current.

ties of three $(TMTSF)_2PF_6$ crystals, all belonging to the same selectively ^{13}C-enriched batch. Only the sites at the center of the TMTSF molecule [C(3) and C(13) in Ref. [13]] have been enriched to 5%. Most of the data presented here have been obtained in a crystal of dimensions $4 \times 0.35 \times 0.25$ mm^3 (mass 550 μg). The cross section is regular all along the sample, but more complex than in smaller crystals, which we ascribe to twinning. For the transport measurements, we trimmed and gold-plated the ends of the crystal, inserted it in a quartz tube holding the NMR coil, and pressed gold contacts against the sample ends by soft iron springs. All the experiments have been performed in the 9.3-T NMR field.

The resistance of the sample decreased monotonically from room temperature to 12 K where the transition to the SDW phase occurs. The residual resistance of the thin gold contact leads has been found to be 1 Ω at 15 K where the sample resistance is very small. All the measurements of the SDW transport have been performed at 4.2 K, well below the transition temperature. Figure 1 shows the conductivity as a function of electric field. The low-field resistance $R_n = 41$ Ω is in agreement with previous magnetoresistance data [14]. A pronounced nonlinearity is observed above a depinning threshold 3.8 mV/cm, close to the lowest values reported in the literature, indicating both a good sample and contact quality.

In Fig. 2, we show voltage noise spectra—obtained by accumulating the Fourier power spectra of time records of the sample voltage—for various dc bias currents above the nonlinear threshold. All the noise power is in a reasonably narrow peak, and, consequently, the mean noise frequency $\overline{\nu}_n$ is well defined. Figure 3 demonstrates that $\overline{\nu}_n$ is proportional to the nonlinear part of the current,

$$I_{\mathrm{SDW}} = I - V/R_n \, . \tag{2}$$

The slight curvature shows that little additional depinning occurs at higher electric fields.

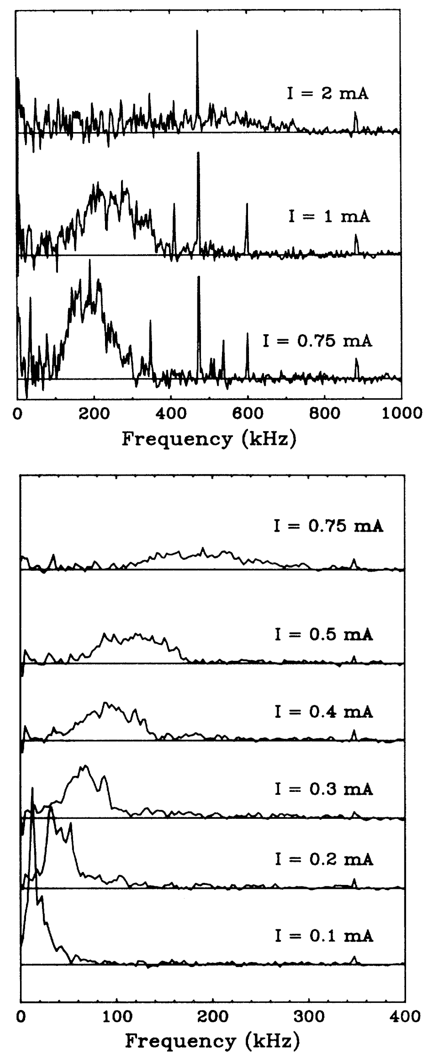

FIG. 2. Conduction noise amplitude as a function of frequency for various dc currents I.

The ^{13}C NMR spectra (Fig. 4) were obtained with a Bruker MSL400 spectrometer at 99.67 MHz. We used a $\pi/2$-τ-π echo sequence with a 2τ=80 μs delay and a 3 s repetition rate, which allows for 70% recovery of the magnetization in the most slowly relaxing part of the spectrum, and checked that no significantly different results are obtained with 1.5 s. We measured the NMR spectra as a function of the dc current passed through the sample. For each spectrum, 12 000 scans were accumulated. Further details on the experimental technique as well as an analysis of the NMR spectrum without sliding and of the spin-lattice relaxation in the same batch can be found in Ref. [3].

At zero current, we observe the U-shaped NMR frequency distributions characteristic of an incommensurate SDW [Fig. 4(b)]. This shape reflects the density

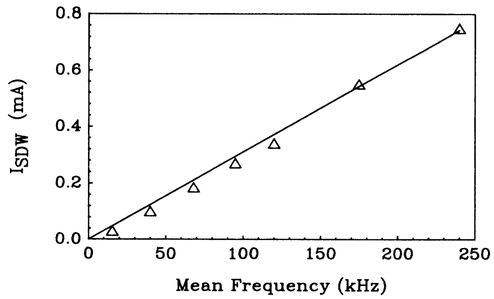

FIG. 3. SDW current vs mean noise frequency. The solid line is a linear fit.

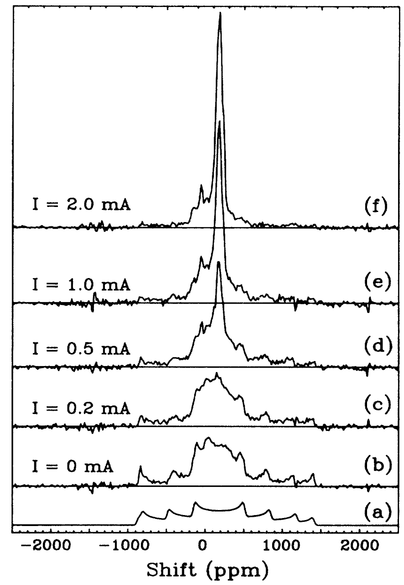

FIG. 4. NMR spectra as a function of the total current I [(b) to (f)], and the decomposition of the $I = 0$ spectrum into incommensurate distributions (a) according to the theoretical line shape calculated in Ref. [3].

of the distribution of the sinusoidally modulated local fields. Only the nuclei at the central carbon sites are observed, because, first, these sites are enriched in ^{13}C, and second, the spin-lattice relaxation times at other sites are much longer. As there are two inequivalent central carbon sites, two distributions with different widths and center frequencies are expected. We observe four distributions, and we attribute this doubling to twinning in the crystal. The decomposition of the spectrum into incommensurate distributions is shown in Fig. 4(a). The four lines have *half* widths $\Delta\nu$ of 110, 98, 65, and 33 kHz, and equal spectral weights. In addition, there is a relatively narrow line with an anomalous shape which cannot be described in a simple plane-wave SDW model, but is clearly associated with the SDW phase as it disappears above the phase transition. The nature of this line, which accounts for about 40% of the NMR intensity, will be further discussed below.

Above threshold, the NMR intensity is gradually transferred from the wings to two overlapping central peaks. In addition, the total intensity initially decreases to 70% of its zero-current value at 0.2 mA and increases back to 96% at 2 mA. Joule heating is ruled out by the fact that the NMR intensity at zero and at the highest current are almost equal. Both features, on the other hand, can be explained by the sliding of the SDW. Assuming a spatially and temporally coherent sliding, one can obtain a closed form for the total NMR intensity, $J_0(4(\Delta\nu/\nu_\phi)\sin^2(\pi\nu_\phi\tau))$, and for the intensity of the narrow central line, $J_0(\Delta\nu/\nu_\phi)J_0(w)$ with $w = (\Delta\nu/\nu_\phi)\sqrt{5-4\cos(2\pi\nu_\phi\tau)}$, where J_0 is the Bessel function [5]. The measured broad noise spectra, however, indicate either a spatial or a temporal incoherence. The sharp features of these spectra suggest that the broadening is mostly due to a spatial distribution of the sliding velocity, rather than temporal incoherence. Assuming $\nu_n = \alpha\nu_\phi$, with either $\alpha = 1$ or 2, and using the measured conduction noise frequency distribution, we calculate the total and central peak intensities of the NMR line, and compare them to the experimental values. For this comparison, we subtract the anomalous line, which is little affected by the sliding (see Fig. 4). We find that the experimental values are different from the calculated ones, which we attribute to the depinning of the SDW in a fraction η of the sample only. From the comparison of the calculated and experimental intensities, we determine the sliding fraction independently from the total and the central peak intensity. From the total intensity, we get 0.45 for $\alpha = 2$ and 0.5 and 0.7 for $\alpha = 1$ at $I = 0.2$ and $I = 0.5$ mA, respectively. From the central peak, we find that at $I = 0.2$ and 0.5 mA, η is about 2 for $\alpha = 2$ and 0.6 for $\alpha = 1$. At $I = 1$ mA, the total intensity is too close to the static value to give an accurate η; from the central peak intensities, we deduce sliding fractions of 1.1 for $\alpha = 2$ and 0.7 for $\alpha = 1$.

Clearly, $\alpha = 2$ cannot account consistently for the data (e.g., we obtain η substantially larger than 1). Similarly any value of α very different from 1 may be discarded. We therefore conclude that the most probable value is $\alpha = 1$. Using this result, we calculate the j_{SDW}/ν_ϕ ratio. The slight increase of sliding fraction we found above is consistent with the curvature of the I_{SDW} vs ν_n plot (Fig. 2), and the ratios we obtain for the various currents lie between 0.85×10^{-19} and 1.0×10^{-19} C, or 30% of the expected $2e$. We believe the SDW current calculated from Eq. (2) may be underestimated, because of a very significant backflow of normal electrons. The backflow of normal electrons in the sliding DW state is well known in CDW's [15], and seems to be very large in the SDW of $(TMTSF)_2PF_6$, as was recently observed in the Hall effect [16].

We find that in SDW's, as in CDW's, the sliding includes a significant fraction (probably all) of the conduction electrons. Since the temperature independent NMR relaxation rate suggests that the SDW effective mass is equal to the band mass [3,17], it is possible that the low frequency microwave response [12] is not due to a simple pinned mode resonance. This resonance may arise from the defects associated with the anomalous NMR line. We have observed this line in several crystals with or without contacts. The fact that it is not narrowed by sliding suggests it could be due to the presence of segregated defects. We attribute the slight intensity decrease of this line with increasing nonlinear current to fluctuations due to the proximity of the sliding parts.

Finally, it was proposed that the conduction noise arises from the interplay of the pinning potential and the periodic DW [18]. For SDW's, the period of the pinning potential is half of the SDW wavelength [19], so that one expects $\alpha = 2$ in this model, in contradiction with our result $\alpha = 1$. Therefore, the voltage modulation probably reflects the creation of a new wave front of the DW out of normal electrons (and the inverse process) [20].

In conclusion, we have shown that in the SDW of $(TMTSF)_2PF_6$, as in CDW's, the nonlinear conductivity is a sliding of the bulk of the DW. The frequency of the voltage oscillations generated by the sliding SDW is equal to the phase winding rate. These results give evidence that the conduction noise is generated by the electron conversion process at the contacts.

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