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Dynamical response of the spin-density-wave mode in tetramethyltetraselenofulvalene hexafluorophosphate [$(TMTSF)_2PF_6$]

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We report frequency-dependent conductivity measurements in the organic conductor tetramethyltetraselenofulvalene hexafluorophosphate [$(TMTSF)_2PF_6$]. The conductivity $\sigma(\omega)$ increases with increasing frequency ω in the spin-density-wave (SDW) state, providing direct evidence for a collective response due to SDW's. A comparison is made with similar observations in charge-density-wave systems.

It is now well established by magnetic measurements[1-3] that the low-temperature semiconducting state of the organic conductor tetramethyltetraselenofulvalene hexafluorophosphate [$(TMTSF)_2PF_6$] results from the development of a spin-density-wave (SDW) ground state at temperature $T_{MI} = 11$ K. In spite of recent efforts to establish whether or not the SDW responds as a collective mode to external ac or dc electric field excitations, the question of collective response remains controversial. Walsh *et al.*[4] reported strongly nonlinear dc conductivity $\sigma(E)$ at small electric field strengths E, and also an ac conductivity $\sigma(\omega)$ measured at microwave frequency (ω) which remained high below T_{MI}, but displayed peculiar dependence on the microwave power. These observations, together with spin resurrection observed by ESR[2,4] were interpreted as a collective response of a pinned spin-density wave, which could be depinned by a sufficiently high electric field. Subsequent experiments,[5] however, showed that a threshold electric field for the onset of dc nonlinearity (which would be expected for a depinning process) did not exist in the SDW state of $(TMTSF)_2PF_6$. In addition, the ac conductivity $\sigma(\omega)$ showed no frequency dependence up to $\omega/2\pi = 100$ MHz, a frequency where strongly ω-dependent response is observed in charge-density-wave (CDW) systems like $NbSe_3$ (Ref. 6) and TaS_3.[7] These observations lead to the conjecture[5] that single particle effects may be responsible for the nonlinear response in $(TMTSF)_2PF_6$. In this Communication we report the observation of frequency-dependent conductivity associated with the SDW phase of $(TMTSF)_2PF_6$. The characteristic frequency where the conductivity starts to be frequency dependent is orders of magnitude smaller than that corresponding to the single particle gap Δ, which is suggestive for a collective mode. Analysis in terms of a pinned mode is also in agreement with a sliding SDW contribution to the dc conductivity at high electric fields. Such analysis leads, however, to strong restoring forces, which is not expected for a weakly pinned SDW.

$\sigma(\omega)$ measurements were performed with an HP 8754A network analyzer, using a helium gas flow system to reduce the length of the coaxial cable between the sample and the network analyzer. Below about 700 MHz the finite cable length was compensated for by a built-in line stretcher, and the experiments can be performed by changing the frequency continuously. Above this frequency a cable length $\lambda/2$, where λ is the wavelength of the excitation signal was chosen for matching and the experiments were performed at each particular frequency as a function of temperature. We have used two-probe configurations to measure $\sigma(\omega)$. Although the contact resistances are comparable with the sample resistance where the conductivity has a maximum at $T \sim 15$ K, several measurements with various ratios between the contact and sample resistances, together with the comparison of $\sigma(T)$ with four-probe measurements,[5] confirmed that in the temperature range considered, the contact resistance is an additional frequency-independent constant R_c which can be subtracted. We have also observed contact capacitance effects which depended on the sample resistance. A careful comparison of the capacitive effects below and above the transition, but for the same sample resistance, confirmed that the dielectric constant ϵ is smaller than 10^6, in agreement with previous observations.[5]

Figure 1 shows $\sigma(\omega)$ measured at selected frequencies at temperatures above and below the metal-insulator transition $T_{MI} = 11.5$ K. The full line represents the experiment performed[4] at 9.1 GHz. We first note that there is no frequency dependence at temperatures well above T_{MI}, and a frequency-dependent response slowly develops somewhat above T_{MI}, with pronounced effects below the transition. This observation, closely analogous to the onset of dc nonlinearity[4,5] rules out any explanation in terms of

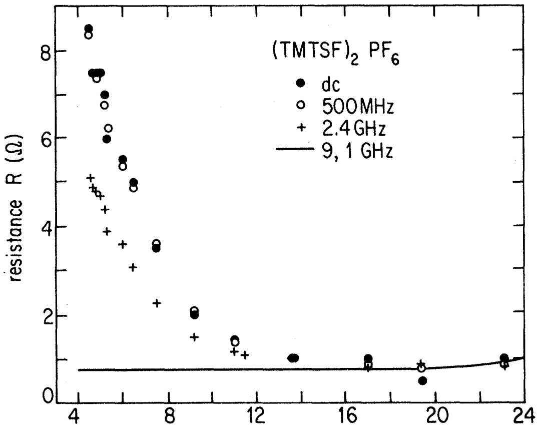

FIG. 1. Temperature dependence of the resistance in (TMTSF)$_2$PF$_6$ at various frequencies. The full line is the result of microwave experiments ($f = 9.1$ GHz), Ref. 4.

microscopic inhomogeneities such as breaks in the sample. If this were the case, ω-dependent response would be observed at all temperatures both above and below the transition.

Figure 2 shows $\sigma(\omega)/\sigma(0)$ as a function of frequency below T_{MI} where $\sigma(0)$ is the dc conductivity. Here we note that the frequency where increased σ is first observed, $\omega/2\pi \sim 10^9$ Hz, is orders of magnitude smaller than the frequency corresponding to the single-particle gap Δ. The dc conductivity below T_{MI} leads to $\Delta/\hbar = 30$ K; this corresponds to a frequency $\omega = \Delta/\hbar = 0.45 \times 10^{12}$ sec^{-1}. Alternatively, if the observed $\sigma(\omega)$ would reflect carrier, or collective mode excitations across a (small) gap, then states would also be thermally populated for $\hbar\omega > k_B T$. We conclude, therefore, that the low frequencies where a frequency-dependent conductivity is observed are in contrast with a simple carrier excitation mechanism.

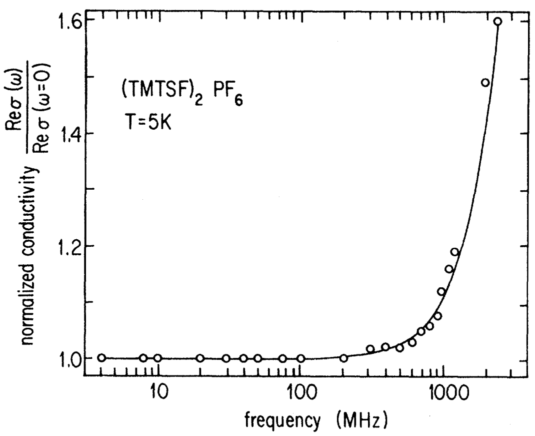

FIG. 2. Frequency-dependent conductivity below the *MI* transition in (TMTSF)$_2$PF$_6$. The full line is Eq. (5) with parameters given in the text.

We proceed therefore to analyze our experiments in the spirit of Lee, Rice, and Anderson,[8] who assume that a pinned collective mode can be described in terms of a classical oscillator in the presence of an alternating deriving force. The complex dielectric constant is given by

$$\epsilon_p(\omega) = \frac{\Omega_p^2}{\omega_p^2 - \omega^2 - i\Gamma\omega} \quad . \tag{1}$$

$\Omega_p^2 = 4\pi n e/m^*$ is the plasma frequency of the mode with n the number of condensed electrons, m^* the effective mass, and Γ the damping constant. $\omega_p^2 = K/m^*$ is the pinning frequency where K is the restoring force. At low frequencies

$$\epsilon(\omega \to 0) = \frac{\Omega_p^2}{\omega_p^2} \tag{2}$$

and

$$\sigma(\omega << \omega_p^2/\Gamma) = \frac{\Omega_p^2}{4\pi(\omega_p^2/\Gamma)^2} \quad . \tag{3}$$

For weakly damped response $\sigma(\omega)$ has a peak at $\omega = \omega_p$, and $\sigma(\omega_p) = \Omega_p^2/4\pi\Gamma$. For a strongly overdamped response ($\Gamma >> \omega_p$) $\sigma(\omega)$ saturates at the value $\sigma(\omega) = \Omega_p^2/4\pi\Gamma$ at frequencies $\omega > \omega_p^2/\Gamma$, and then falls off at frequencies $\omega > \omega_p$. At high electric fields when the SDW condensate slides freely and there is no restoring force, the dc conductivity is given by

$$\sigma(E \to \infty) = \frac{\Omega_p^2}{4\pi\Gamma} \quad , \tag{4}$$

Eqs. (3) and (4) can then be combined to give the low-frequency conductivity

$$\frac{\sigma(\omega << \omega_p^2/\Gamma)}{\sigma(E \to \infty)} = \frac{\omega^2}{(\omega_p^2/\Gamma)^2} \quad . \tag{5}$$

The full line in Fig. 2 is a fit of the experimental points to Eq. (5) with $\sigma(E \to \infty)$ taken as the conductivity before the transition.[4] This leads to $\omega_p^2/\Gamma = 5 \times 10^9$ sec^{-1}. For a weakly damped response ($\omega_p > \Gamma$) this would imply a maximum in $\sigma(\omega)$ below this frequency with $\sigma(\omega)$ which strongly decreases at frequencies both above and below ω_p. The conductivity measured at microwave frequency $\omega = (2\pi)9.1 \times 10^9$ sec^{-1} $= 10^{10}$ sec^{-1}, however, is the same as the high-field conductivity,[4] suggesting strongly that the system is strongly damped, $\Gamma > \omega_p$ and $\omega_p >> 6 \times 10^{10}$ sec^{-1}. A similar analysis leads to the same conclusion for the CDW systems NbSe$_3$ (Ref. 6) and TaS$_3$.[9]

Although our analysis in terms of a pinned SDW mode leads to a self-consistent interpretation, the observed parameters are surprising when compared with those obtained in pinned CDW systems.[6,7] Both $NbSe_3$ and TaS_3 would seem to have a stronger collective mode pinning than $(TMTSF)_2PF_6$. A stronger pinning is expected for $NbSe_3$, where impurities are responsible for CDW pinning, since impurities should provide stronger pinning interactions with CDW's than with SDW's. The high transition temperature of TaS_3 would imply a stronger pinning for commensurate modes. Another factor arguing for weak pinning in $(TMTSF)_2PF_6$ is the small SDW amplitude,[10] observed by NMR. Although the large characteristic frequency can be explained by a small SDW mass, the observed dielectric constant ϵ cannot.

The dielectric constant

$$\epsilon(\omega \rightarrow 0) = \frac{4\pi ne^2}{K}$$

reflects only the restoring force $K = m\omega_0^2$ acting on the pinned mode. In $(TMTSF)_2PF_6$, ϵ is two orders of magnitude smaller than that in $NbSe_3$ or TaS_3. In $NbSe_3$ $\epsilon = 2 \times 10^8$ below the second CDW transition, and $\epsilon = 10^7$ in TaS_3 in the CDW state, while in $(TMTSF)_2PF_6$ $\epsilon = 10^6$. Since these three compounds have roughly the same number of condensed electrons, this would imply a much larger restoring force for SDW pinning than for CDW pinning, contrary to the previous expectation.

In conclusion, our experiments suggest a description of SDW dynamics in terms of an overdamped response, and are in clear conflict with interpretations of carrier excitations across a single-particle gap. The observed frequency dependence points to two important questions. (1) Why is the restoring force for SDW pinning much larger than for CDW pinning? (2) What is the source of the observed strong nonlinearity in $(TMTSF)_2PF_6$?

The first question may be answered by systematic studies involving materials with various impurity concentrations and also by further experiments at higher (optical) frequencies. Concerning the second question, hot-electron effects were already suggested[11] to account for $\sigma(E)$. The frequency dependence would, in this case come from the existence of barriers in the semiconducting state. ac-dc coupling experiments, similar to those performed[12] in $NbSe_3$ may clarify this point.

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