Effect of spin-density wave in $(TMTSeF)_2PF_6$ on dc and microwave conductivity

L. I. Buravov, V. N. Laukhin, and A. G. Khomenko

Division of Institute of Chemical Physics, USSR Academy of Sciences
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The temperature dependences of the conductivities of the salt $(TMTSeF)_2PF_6$ for direct current and at 10^{10} Hz were investigated. It is shown that an additional contribution to the microwave conductivity appears when a spin-density wave is produced.

The organic conductor[1] $(TMTSeF)_2PF_6$, which becomes a superconductor with $T_c \sim 1$ K under pressure (Ref. 2), has at normal pressure metallic conductivity above the temperature $T_s = 11.5$ K (Ref. 1) at which it undergoes a metal-insulator transition accompanied by formation of a spin-density wave.[3]

An unusual behavior of the microwave conductivity of this conductor was observed in Refs. 4–6 below the point T_s, where $\sigma_{\rm micr}$ exceeds greatly the dc conductivity. It is proposed that this effect is due to the presence of a spin-density wave. The values cited for this effect in Refs. 4 and 6, however, differ noticeably. A decrease in the resistance of the crystals at $T < T_s$ is observed also at higher dc densities[4]; this decrease may be due to some heating of the samples. We have undertaken here a more detailed study of the behavior of the resistance of the $(TMTSeF)_2PF_6$ crystals as the current density is varied, as well as at 10^{10} Hz.

MEASUREMENT PROCEDURE

The crystals investigated were grown by an electrochemical method close to that described in Refs. 1 and 7. The crystal dimensions reached $7\times0.1\times0.1$ mm. To measure the dc conductivity by the four-point method, gold stripes were sputtered on the crystal, which was then clamped to platinum wires of 30 μm diameter (Fig. 1). This way of mounting the crystals yielded low-resistance electric contacts and prevented cracking of the samples after several cooling and heating cycles.

The microwave conductivity was measured at 10^{10} Hz by a somewhat more refined procedure[8] with periodic passage of the generator frequency through resonance. The increase of the width of the cavity passband following placement of the sample in the antinode of the electric field was determined from the change of the maximum of the coefficient of the microwave-signal transmission through the cavity. Small changes of the signal were measured with a pulse-height discriminator that cut off the lower part of the resonance-pulse curve and left at the pulse peak a small part that was amplified and recorded from the oscilloscope screen. With a sufficiently strong signal to the detector (≈ 20 mV) it was possible to detect by this procedure, using a precision microwave attenuator, small signal changes ($\gtrsim 0.01$ dB).

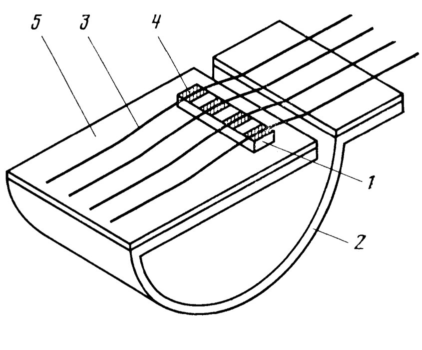

FIG. 1. Mounting and wiring and crystal: 1—crystal, 2—copper foil, 3—platinum-wire electrodes, 4—gold stripes, 5—insulating layer.

It must be noted that the insertion of a long sample in the cavity changes the coupling between the cavity and the waveguides, thereby adding to the change of the transmission coefficient, besides the losses in the sample itself. Experiments with copper samples, which introduced negligible losses into the cavity, have shown that this additional change (in decibels) is proportional to the sample-induced shift of the resonant frequency; the proportionality coefficient has a definite dependence on the cavity temperature but not on the shape or size of the sample in a rather wide range. This temperature dependence of the proportionality coefficient was determined beforehand, and when the microwave conductivity was subsequently measured the change introduced in the transmission coefficient by the change of the coupling constants was taken into account in all the experiments.

The microwave conductivity was calculated using the results of Ref. 9 with allowance for the skin effect; these results were obtained under the assumption that the coupling coefficient of the cavity with the waveguides was constant. It follows from Ref. 9 that for a conducting round cylinder of radius R, placed in the cavity at the antinode of the electric field E and parallel to the field, we have

$$\sigma=(2R^2\omega\mu_0)^{-1}[1+4\pi^2\delta^2R^2/\alpha\Delta\lambda^2]^2 \quad \text{for } R\geqslant h, \tag{1}$$

where h is the skin-layer thickness, σ is the sample conductivity, ω is the cavity frequency, μ_0 is the magnetic permeability of vacuum, δ is the relative shift of the cavity frequency, α is the filling factor, λ is the wavelength in the vacuum, Q is the cavity figure of merit, and Δ is the change of Q^{-1} following introduction of the sample in the cavity. For a sample with rectangular cross section, R is Eq. (1) is replaced by $\widetilde{R} = ab/(a+b)$, were ab is the cross section of the sample. At low temperatures, the microwave power fed to the cavity

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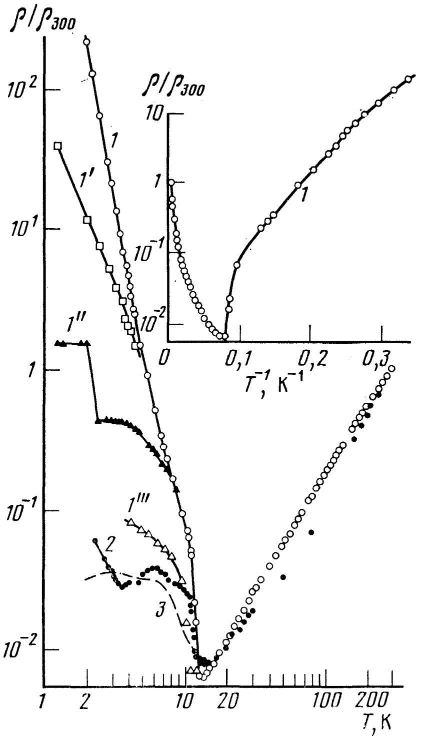

FIG. 2. Temperature dependences of the relative resistivity for different values of direct and microwave currents: 1—$I = 1$ μA 1'—$I = 1$ mA, 1''—$I = 10$ mA, 1'''—$I = 30$ mA, 2—10 GHz, 3—9.1 GHz (results of Ref. 6).

is 200 μW, corresponding to a maximum electric field strength $E_m \approx 10$ V/cm outside the sample at the antinode.

MEASUREMENT RESULTS

The dc conductivity was measured in a wide range of working currents from 1 μA to 40 mA, corresponding to current densities in the sample from 10^{-2} to $4 \cdot 10^2$ A/cm^2. Figure 2 shows a typical temperature dependence of the relative resistance at various dc currents. It can be seen that, just as in Ref. 1, the resistance minimum is observed in the region of $T = 13$ K. In this case $\rho_{300}/\rho_{13} = 130$–160, depending on the cooling cycle, and is practically independent of the working current ($\rho_{300} \approx 2 \cdot 10^{-3}\ \Omega \cdot$ cm). We note that $\rho \propto T^{1.7}$ at $T > 13$ K. The resistance increases abruptly near 12 K as a result of the metal-insulator transition. With further lowering of the temperature the resistance measured at $I = 1$ μA dc, increases rapidly, with an activation energy ≈ 25 K, in the temperature interval 3–10 K (curve 1 and inset of Fig. 2). When the current through the sample is increased, however, the resistance growth slows down greatly (Fig. 2). Thus, whereas $\rho_{1,3}/\rho_{300} \approx 10^3$ at $I = 1$ μA, we have $\rho_{1,3}/\rho_{300} \approx 40$ at $I = 1$ mA.

One cannot exclude a possible connection between this effect and the nonlinearity of the current-voltage characteristic of the crystal, a nonlinearity due to cessation of the spin-density wave. The sample resistance is lowered also by heating. This is clearly observed when the current is increased. At certain currents we have observed resistance jumps, both upward in the vicinity of ~4.2 and 2.2 K, and downward at intermediate temperatures. An example of such a jump at $T \approx 2.2$ K is shown on curve 1'' of Fig. 2. Obviously, these jumps are due to changes of the conditions of heat transfer from the sample, inasmuch as at 4.2 K the sample is dropped into the bath with liquid He^4, and the latter becomes in turn superfluid at $T < 2.2$ K; at $2.2 < T < 4.2$ K a gas bubble can be produced in the liquid-He^4 bath and affect the heat transfer adversely. At $I = 30$ mA there is practically no dielectric growth of the resistance and the minimum of the curve is shifted downward somewhat in temperature, likewise indicating that the sample is heated.

Figure 3 shows the dependences of the sample resistance on the current temperatures 1.3 and 4.2 K. It can be seen that the section on which the temperature is independent of current is smaller the lower the temperature. Generally speaking, this behavior can be due to heating of the sample. Indeed, it is easier to heat the sample at lower temperature, and the spin-density wave is easiest to shut off the higher the temperature and the smaller the gap produced in the metal-insulator transition. It must be noted here, however, that at $T = 1.3$ K the electric field in the sample is larger by more than two orders than at $T = 4.2$ K if $I \leqslant 2$ μA, since the sample resistance increases rapidly with decreasing temperature. Therefore the shutoff of the spin density wave and the corresponding increase of the conductivity at $T = 1.3$ K can be observed even at such low currents.

In the microwave measurements we have observed that the resistivity at 10^{10} is close to the dc resistivity in the interval 1.2–300 K, but at $T < 10$ K the microwave resistivity is substantially lower than the dc resistivity at $I = 1$ μA (Fig. 2). Our results for the temperature dependence of $\rho_{\rm micr}(T)$ agree qualitatively with the data of Ref. 6 (curve 3 of Fig. 1), obtained for the interval 2–22 K at 9.1 GHz.

With allowance for the skin effect, the microwave electric field on the sample surface at $T \approx 5$ K was $E_k = \omega\varepsilon_0 \widetilde{R} E_m/\sqrt{2} n\sigma h \approx 0.03$ V/cm (Ref. 9), where ε_0 is the dielectric constant of the vacuum and n is the sample-depolarization coefficient. The power absorbed by the sample, with allowance for the fast passage of the generator frequency through the resonance, is 0.5 μV. This absorbed power is

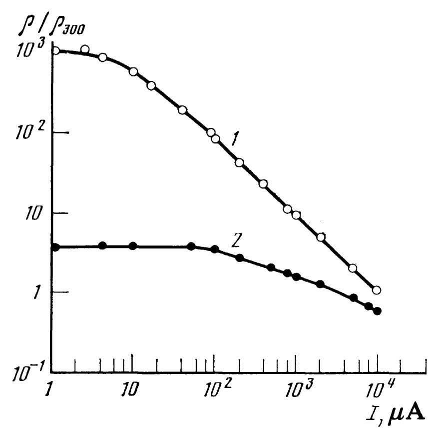

FIG. 3. Current dependence of the relative resistivity: 1—$T = 1.3$ K, 2—4.2 K.

comparable with the result of dc measurements at $I = 100$ μA, if account is taken of the resistance of the current contacts of the sample. At $T < 10$ K, ρ_{micr} turned out to be independent of the microwave electric field when the latter was decreased by a factor of four. In this case the microwave electric field on the sample surface becomes comparable with the dc field in the measurements at $I \lesssim 100\,\mu$A. It must thus be assumed that the observed difference between ρ_{micr} and ρ_{dc} for small currents at $T < 10$ K is a property of the material and is not due to heating or to the action of strong microwave fields.

DISCUSSION

The contribution of a collective mode (charge-density or spin-density wave) to the complex ac admittance was calculated in Ref. 10 for an ideal case without friction or pinning. If these factors are taken into account, the motion of the collective mode in a weak electric field can be written in the form of the equation of motion of a classical oscillator[5]:

$$m_c\ddot{x}+\alpha_c\dot{x}+Kx=e_0E_0e^{-i\omega t}, \qquad (2)$$

where m_c is the effective mass of the condensed electrons, α_c is the friction coefficient, K_x is the restoring force of the pinning center, and E_0 is the amplitude of the field inside the conductor. It follows from (2) that the collective-mode contribution to the ac resistivity is[5]

$$\sigma_c(\omega) = \mathrm{Re}\left(\frac{n_c e_0 \dot{x}}{E}\right) = \frac{\varepsilon_0\Omega_p{}^2\Gamma\omega^2}{(\omega^2_{pin}-\omega^2)^2+\Gamma^2\omega^2}, \qquad (3)$$

where

$$\omega_{pin}=\left(\frac{K}{m_c}\right)^{1/2}, \quad \Gamma=\frac{\alpha_c}{m_c}, \quad \Omega_p{}^2=\frac{n_c e_0{}^2}{m_c\varepsilon_0},$$

n_c is the number of condensed electrons, and e_0 is the charge of the electron. The contribution $\sigma_c(\omega)$ has a maximum $\varepsilon_0\Omega_p{}^2/\Gamma$ at $\omega = \omega_{pin}$ and vanishes as $\omega \to 0$ or $T \to T_s$. The contribution to the conductivity from the free carriers at $T < T_s$ at the frequency ω is $\sigma_1(\omega) \approx \sigma_{dc}$, so long as $\omega \ll \tau^{-1}$, where τ is the relaxation time. The ratio of the total measured conductivity to the dc conductivity in this frequency region is therefore

$$\frac{\sigma_\Sigma(\omega)}{\sigma_{dc}} = \frac{\sigma_c(\omega)+\sigma_1(\omega)}{\sigma_{dc}} = 1+\frac{\varepsilon_0\Omega^2\Gamma\omega^2/\sigma_{dc}}{(\omega^2_{pin}-\omega^2)^2+\Gamma^2\omega^2}. \qquad (4)$$

It has been assumed here that it is precisely that the contribution $\sigma_c(\omega)$ that causes the difference between the dc and microwave resistances at $T < T_s$. A similar conclusion was reached earlier in Refs. 5 and 6.

The value of Ω_p for $(TMTSeF)_2PF_6$ was determined in Ref. 11 from optical measurements of the reflection coefficient, viz., $\Omega_p \approx 2.2 \cdot 10^{15}\ s^{-1}$ in the interval 25–300 K; it apparently remains unchanged at $T \ll T_s$, since the gap is $E_g \ll \hbar\,\Omega_p$ and $m_c = m$ for the spin-density wave,[10] where m is the effective conduction-band carrier mass. The values of Γ and ω_{pin} can be estimated from the experimental results. We note in this connection that, first, the ratio $\sigma_\Sigma(\omega)/\sigma_{dc}$ is known for $\omega_1 = 2\pi \cdot 10^{10}\ s^{-1}$ from the present paper (≈ 40 at $T = 5$ K); second, it was shown experimentally in Ref. 5 that at low frequencies $\sigma_\Sigma(\omega)/\sigma_{dc} = 1 + \omega^2/A^2$, where $A \approx 2 \cdot 10^{10}\ s^{-1}$ at $T = 5$ K. From this and from Eq. (4) we obtain two equations with two unknowns, and ω_{pin}, for $T = 5$. Solution of these equation yields $\omega_{pin} \approx 10^{11}\ s^{-1}$ and $\Gamma \approx 10^8\ s^{-1}$. We note that no assumptions were made here concerning the values of Γ and ω_{pin}, in contrast to Ref. 5 where it was assumed beforehand that $\Gamma > \omega_{pin}$.

It is is assumed that at $T < T_s$ the resistivity at the frequency ω_{pin} has a temperature dependence of the metallic type: $\rho(\omega_{pin}, T) = \rho_0 + \rho_1 T^k$, where ρ_0 is the residual resistivity and $k > 1$, we can estimate the temperature dependence of the resistance in this interval for the frequency $f \approx 10$ Hz. Recognizing that $(2\pi f)^2 \ll \omega^2_{pin}$, we obtain from (3)

$$\rho(f, T) \propto [\rho(\omega_{pin}, T)]^{-1} \propto (\rho_0+\rho_1 T^k)^{-1}. \qquad (5)$$

It is possible that some general increase of ρ_{micr} as the temperature is lowered at $T < 10$ K is due to the fact that $\rho_1 T^k$ remains comparable with ρ_0; the nonmonotonic variation of $\rho_{micr}(T)$ in the interval 3–6 K remains unexplained, however, in view of the temperature dependence (5).

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