Nonlinear microwave properties of quasi-one-dimensional $(TTT)_2I_3$ crystals

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It has been found experimentally that the microwave admittance of the tetrathiotetracene iodide $[(TTT)_2I_3]$ crystal varies with the magnitude of a direct current flowing through the crystal ($f = 40$ GHz, $T = 300$ K). Two processes have been distinguished: a slow process related to the heating of the crystal and a fast process with $\tau < 10^{-7}$ s. The occurrence of this fast process is at odds with the conventional theory for transport processes in organic quasi-one-dimensional metals.

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No electrical nonlinearity has been observed in quasi-one-dimensional metals at temperatures above the Peierls transition, except in the obvious case of a change in the admittance due to a heating of a crystal by a current flowing through it.[1] The theoretical model of Lenahan and Rowland[2] predicts that quasi-one-dimensional metals should not exhibit a nonlinear admittance at temperatures above the semiconductor-metal transition. A nonlinearity may be observed below the transition temperature, as has been confirmed experimentally.[2–4]

We are reporting here a study of the application of electromagnetic radiation at the frequency 4×10^{10} Hz to a crystal of the organic metal tetrathiotetracene iodide[1)] of the composition $(TTT)_2 I_3$ ($\sigma \approx 10^3$ mho/cm at $T = 300$ K). We measured the reflection coefficient of the sample during the flow of a low-frequency sinusoidal current through it. The reflection coefficient is unambiguously related to the microwave admittance of a sample. The contacts through which the low-frequency current was supplied to the sample were placed outside the region in which the microwave field interacted with the crystal, so that they would not affect the process under study. All the measurements were carried out at room temperature.

The experimental arrangement is shown in Fig. 1. The sample (1) lies midway across the broad wall of a waveguide and is oriented parallel to the electric field

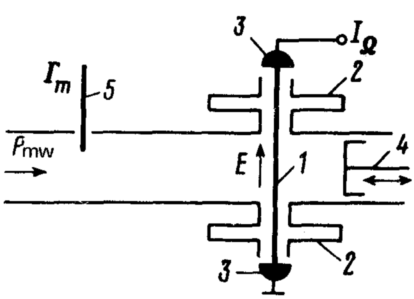

FIG. 1.

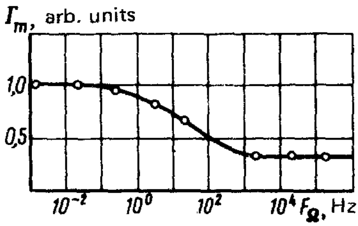

FIG. 2.

vector. Quarter-wave short-circuiting radial resonators 2 provide open-circuit conditions for the microwave current induced in the sample at the points at which the resonators are attached. The low-frequency current enters the sample through contacts 3. The movable short-circuiting plunger 4 is used along with adjustable rods to provide the necessary matching. The reflection coefficient is measured with measuring-line probe 5.

The sample was a $(TTT)_2 I_3$ crystal 1 cm long with a cross-sectional area of $30 \times 60\ \mu m^2$ and with aquadag contacts. The cw microwave power level did not exceed 2 mW; the peak value of the sinusoidal current through the sample did not exceed 5 mA.

Figure 2 shows the measured index of the modulation of the reflection coefficient, Γ_m, plotted as a function of the frequency of the sinusoidal current through the sample. It can be seen from this figure that there are two processes which determine the change in the microwave admittance of the sample as a low-frequency current flows through it. The slow process is very probably a trivial manifestation of the heating of the crystal. In measurements of the dc conductivity of the samples by a four-probe method, we observed a deviation from a linear voltage-current charcteristic; this nonlinearity disappeared in pulsed operation, under conditions such that the crystal was not heated.

The fast process is manifested at frequencies above 10^3 Hz, and its intensity remains constant up to 10^6 Hz. To learn more about this fast process, we supplied a pulsed current to the sample in place of the sinusoidal current. Figure 3 shows oscilloscope traces of a current pulse 0.6 μs long which passed through the crystal (the

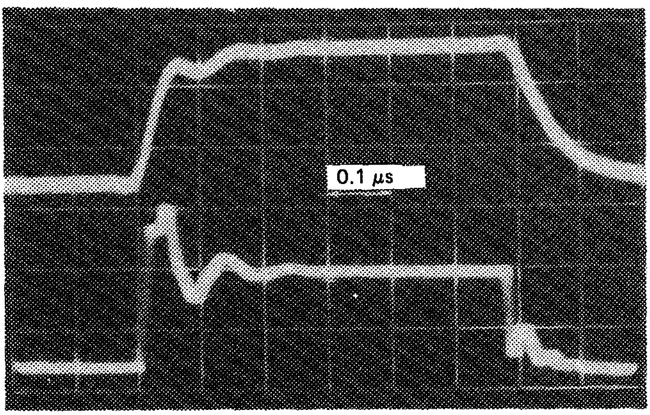

FIG. 3.

lower trace) and the signal from the measuring-line probe. This signal is a measure of the change in the reflection coefficient caused by the current flowing through the crystal. The observed distortion of the pulse shape results primarily from the finite response time of the measuring line. It can be seen from this trace that the time constant of the fast process is less than 10^{-7} s.

In summary, these experiments reveal that a sinusoidal or pulsed current flowing through a $(TTT)_2 I_3$ sample affects the microwave admittance of the sample at room temperature. This result has not been predicted theoretically, and it has not previously been observed experimentally. The short response time of this process implies that the nonlinearity is of an electronic nature.

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[1)]The crystals were grown by É. B. Yagubskiĭ and G. I. Zvereva.

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