Cooperative transport phenomena in phase-separated degenerate antiferromagnetic semiconductors

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The response to a strong electric field of degenerate antiferromagnetic semiconductors EuTe, which are spontaneously separated into antiferromagnetic and ferromagnetic phases, is investigated. While in weak fields their response is ohmic, in strong fields a sequence of very high peaks appears against the ohmic-type background, beginning at a certain threshold field strength. Their spacing decreases with increasing field strength. These peaks can be explained by the cooperative motion of charged ferromagnetic microregions inside the antiferromagnetic host.

1. Introduction

The phenomenon of phase separation can be realized in degenerate antiferromagnetic (AFM) semiconductors which exhibit high-temperature superconductivity and those which do not exhibit it. It was first predicted and investigated theoretically twenty years ago in Refs. 1 and 2 (a detailed description of their results is given in Refs. 3 and 4. The phase separation was subsequently observed in nonsuperconducting EuTe (Refs. 5 and 6) and EuSe (Ref. 7) (see an analysis of these experimental data in Refs. 3 and 4). Recently phase separation was observed in superconducting systems based on AFM semiconductors (e.g., Ref. 8) The problem of phase separation in such systems has also attracted the attention of theoreticians (e.g., Refs. 9 and 10).

In the present paper we describe a new effect which occurs in phase-separated AFM semiconductors EuTe: the appearance of high-current peaks against the ohmic-type background in strong electric fields. These peaks form a periodic sequence, whose period decreases with increasing field strength. This phenomenon can be explained by the cooperative motion of charged microregions of the ferromagnetic (FM) phase which appears in the crystal as a result of its separation into AFM and FM phases.

2. Experimental data

Nonstoichiometric single crystals of EuTe with excess of Eu were studied. Samples were grown by the chemical transport method in hermetically sealed Mo crucibles at 2000 °C. The deficiency of Te was determined from the temperature and exposition time. The length of sample varied from 1.2 to 3 mm, and its cross section was 1 mm×1 mm. As for its electric properties, at liquid-nitrogen temperatures it behaves like a conven-

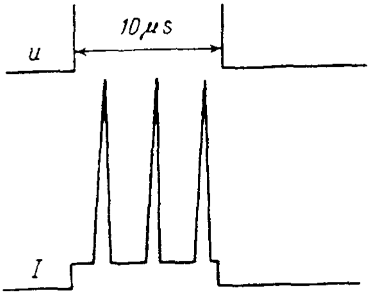

FIG. 1. Time dependence of the voltage U and the current I.

tional n-type degenerate semiconductor. Its conductivity is on the order of 100 S/cm and the conduction electron density is on the order of 10^{19} cm^{-3}. (Such properties exhibit EuTe crystals doped by I.) With a decrease in the temperature, however, the conductivity of the test samples decreases sharply by several orders of magnitude (sometimes by a factor greater than 10^6), reaching at 4.2 K values typical of nondegenerate semiconductors. Such a drop was observed earlier in Refs. 5 and 6. Magnetic measurements carried out in those papers confirm the coexistence of AFM and FM phases in the low-temperature region, which means that this drop in the conductivity is a consequence of phase separation (see Refs. 3 and 4).

To avoid a heating of the samples, we used an impulse regime. Rectangular voltage pulses of 10-μsec duration were applied to the samples at 4.2 K; the pulse frequency was in the range 1–10 Hz. Most interesting are properties of samples with a resistivity on the order of 10^3 Ω·cm at 4.2 K. Below we describe in detail the properties of a sample with a resistivity of 1.9×10^3 Ω·cm at 4.2 K. The threshold value of the field strength is about 1.4 kV/cm; the response of the system reproduces the shape of the voltage pulse. But after reaching the threshold field, a triangular peak appears at the trailing edge of the current pulse. The height of the peak is larger than the height of the rectangular background by a factor of 10–20. The duration of increase and decrease of the current in the peak was found to be independent of the field applied in the field range investigated. This duration is 0.7 μsec. On further increases of the applied voltage, the current peak shifts toward the leading edge of the pulse without changing its shape. Then another peak appears at the trailing edge, which also shifts toward the leading edge with increasing voltage. The third peak then appears at the trailing edge, and so on (Fig. 1). The maximum number of the peaks observed reaches 5. Thus, the peak spacing t decreases with increasing voltage (Fig. 2).

An additional information may be obtained from the current-voltage characteristics of the sample recorded at the trailing edge of the current pulse with a strobe duration of 50 ns (Fig. 3). Below the threshold field at which the current peak appears for the first time, the height of the rectangular current pulse approximately corresponds to the Ohm's law (more precisely, the current dependence on the voltage is superlinear). As can be seen from this figure, after the current peak appears at the threshold voltage, this peak first disappears and then appears again. Disappearance of the peak at the trailing edge is a consequence of its shift toward the leading edge. All the current peaks are of the same height, but their spacing depends on the voltage which decreases with the field. In con-

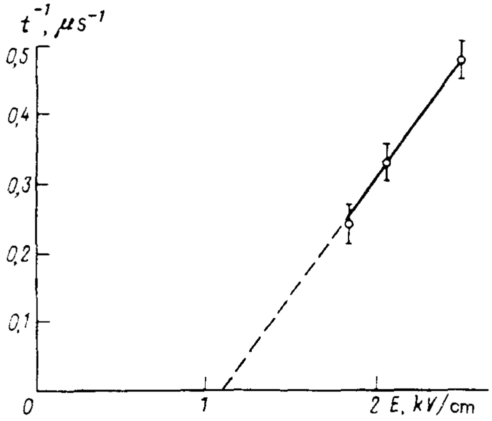

FIG. 2. Dependence of the current peak spacing t (μsec) in the multi-peaked response to the applied field E (kv/cm).

trast, the peak width increases with the field. This occurs because the field depends on the rate at which the peak moves along the current pulse.

Comparison of the results presented above with those for samples with resistivities ρ of 2×10^3 and 3.5×10^3 Ω·cm at 4.2 K shows no evidence of a ρ-dependence for the duration of the current pulse. The relative pulse height seems to decrease with increasing ρ. In the sample investigated the current peaks disappear at 8 K, i.e., still below the Néel temperature, 9.6 K. They are absent at any temperatures for samples with resistivities, at 4.2 K, lower than 10 Ω·cm and higher than 10^5 Ω·cm.

3. Theoretical interpretation

A natural way to explain these experimental results is to attribute the rectangular current pulses to the charge transport by the conduction electrons whose density is de-

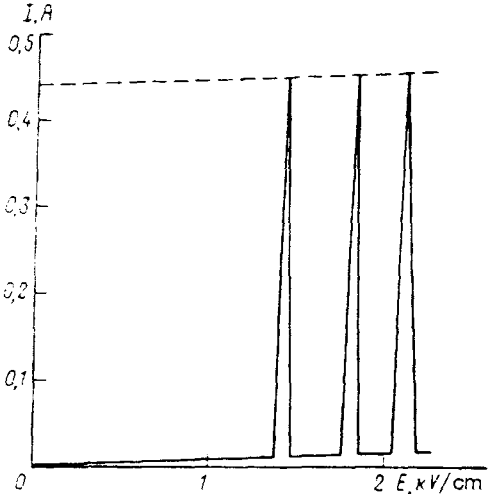

FIG. 3. Current I (A) versus applied field E=(kV/cm) with the strobe fixed at the trailing edge of the current pulse.

tectable at 4.2 K in the samples with current peaks (from 10^{13} to 10^{14} cm^{-3}). As for the current peaks, they can be explained by the cooperative motion of the negatively charged FM microregions that exist in the EuTe crystals. To clarify this point, it is instructive to give a brief review of the results.[1,2]

According to these results, the phase separation in degenerate AFM semiconductors is directly related to the ferron state in the nondegenerate semiconductors which were first introduced in Ref. 11: A charge carrier creates a FM region inside the AFM host and makes it stable by localization in it. The ferron state corresponds to the phase separation on the scale of a single carrier. In degenerate semiconductors the phase separation is meant to be a cooperative ferron state in which the Coulomb interaction plays an important part. Thus, in contrast with the phase separation in the first-order phase transition, here the separation of the crystal into two regions, one of which is the FM and the other the AFM, is impossible since the former would contain all the charge carriers and the latter be devoid of them. Since the FM and AFM portions of the crystal are charged oppositely, the Coulomb energy of the system may be reduced if the FM and AFM regions alternate.

It is very important that the set of AFM and FM regions should form a united system due to the Coulomb interaction between them. If the carrier density is not very large, this property manifests itself in a periodic arrangement of FM regions inside the AFM host. Typically, each FM region may contain up to several tens of electrons, and its radius is on the order of several nanometers. With increasing carrier density n, the FM portion of the crystal converts from a multiply connected region to a singly connected region which corresponds to AFM droplets inside the FM host.

Only the former case will be considered here since it corresponds to the insulating state of the crystal at $T=0$, while the latter case corresponds to a high-conducting state. The set of FM droplets cannot move freely throughout the crystal since they are pinned by spatial fluctuations of the electrostatic impurity potential inherent in degenerate semiconductors (they disturb the periodic arrangement of the FM droplets). On the other hand, the electrons of each droplet are locked inside it and cannot take part in the charge transport. At finite temperatures, the crystal behaves like a nondegenerate semiconductor since free electrons appear in the conduction band of the AFM portion of the crystal.

In strong fields, however, depinning of the FM droplets does occur, and they are able to move along the crystal as a single entity, since they are connected with each other by Coulomb forces. Thus, a cooperative charge transport becomes possible in systems with charge density waves, although its characteristic features are different. Here they are determined by the fact that in the field the FM droplets retain their periodic arrangement. Since the external field creates a preferred direction, this structure may be thought of as consisting of droplet crystalline layers perpendicular to this field. These layers move to the cathode and when they reach it, a current peak appears. But the current of droplets vanishes if the cathode borders with an insulating AFM layer situated between the droplet layers. Thus, as a function of time, the droplet current displays peaks, between which it vanishes, in agreement with the experimental data described above.

To make sure that this explanation does not contradict our experimental data, we should compare the theoretical values of the volumes V_F for the ferromagnetic droplets with values of the volume per droplet, V_p, estimated from experimental data. According to Ref. 2, the theoretical radius of droplets amounts to 1–3 nm; i.e., V_F is between 3×10^{-20} and 10^{-19} cm^{-3}. The theoretical number of conduction electrons in each droplet N reaches several tens.

In order to estimate V_p, we note that according to the experimental values of the height (0.45 A, Fig. 3) and duration of the current peak (0.7 μsec), the number of conduction electrons producing this peak amounts to about 10^{12}. With the total sample cross section of 1 mm^2 and N between 10 and 50, this value corresponds to the cross-sectional area per droplet in the range 10^{-13}–(5×10^{-13}) cm^2. The volume per particle, V_p, should therefore be between 3×10^{-20} and 4×10^{-19} cm^3.

This volume includes the ferromagnetic droplet and its antiferromagnetic environment. In the geometry considered here the ferromagnetic droplets which are separated from each other can exist if their volume V_F is less than half of the volume per droplet V_p. Obviously, the estimates obtained above satisfy this condition.

The specific feature of the cooperative transport under discussion is the fact that it is realized via the hopping motion of droplets. One might think that the difference in the pinning potentials for different droplets should lead to an incoherence in their motion which would manifest itself in the spreading of the current peaks. It should be kept in mind, however, that the interaction between droplets may help to surmount the pinning potentials simultaneously by all the droplets. The system of the FM droplets should respond to a certain average pinning potential, which would prevent the current peaks from spreading. In addition, it can be shown that the correlations between the droplets may increase the probability of hopping.

To illustrate the latter statement, we will analyze a simplified model. A pair of particles that model the FM droplets is initially located on a pair of neighboring sites A and B. The particles then go over from site A to site C and from site B to site D, respectively, where sites C and D are neighbors. The Hamiltonian of the system is'

$$H=H_0+H_1\ , \tag{1}$$

$$H_0=\sum E(g)n_g+1/2\sum U(g,g')n_gn_{g'}\ , \tag{2}$$

$$H_1=B\sum a_g^{\dagger}a_{g'}\ . \tag{3}$$

The commutation relations between the operators $a_g^{\dagger}$ and a_g of a particle located on the site g may be arbitrary, $n_g=a_g^{\dagger}a_g$. In what follows we set

$$E_A=E_B=0,\quad E_C=E_D=K; \tag{4}$$

$$U_{AB}=U_{CD}=0,\quad U_{AD}=U_{BC}=U. \tag{5}$$

Considering H_0 as the zeroth-order Hamiltonian and H_1 as the perturbation, we obtain the following expressions for the probability $W_{AB,CD}$ for a joint transition of a pair of particles from atoms A and B to atoms C and D at a large enough time t:

$$W_{AB,CD}=W_{AC}W_{BD}\ , \tag{6}$$

$$W_{AC}=W_{BD}=(2\pi B^2 t/\hbar)\delta(K) \tag{7}$$

for the case of noninteracting particles ($U=0$);

$$W_{AB,CD}=(2\pi B^4 t/U^2\hbar)[2\delta(K)+\delta(K-U)+\delta(K+U)] \tag{8}$$

for the case of interacting particles.

In more sophisticated models, in which the interaction of FM droplets with phonons is taken into account (as in Ref. 12, where the phonon-assisted hopping of the ferron is treated), the energy spectrum of the system should be continuous. Thus, the δ-functions in (6) and (8) should be replaced by an effective density of states $\rho(E)$, which is a continuous function of the energy and which should peak at $E=0$.

As can be seen from a comparison of (6) and (8), the intersite hopping of a particle, which does not interact with another particle, is forbidden at $K\neq 0$. Interaction with another particle, however, may make their joint transition allowed if $K=U$. We note that the transition probability $W_{AB,CD}$ for the uncorrelated particles (6) is the product of the transition probabilities for single particles and for this reason is proportional to t^2. But the transition probability (8) for correlated particles is proportional to t, like for a single particle. This means that correlated particles move jointly like a united particle, which was assumed in the analysis presented above. This effect is independent of the sign of U. It should be kept in mind, however, that such a motion is possible if only the interparticle correlations are not too strong. In fact, even if the δ-functions are replaced by $\rho(E)$ in (8), the probability should vanish as $1/U^2$ with increasing U.

Other possible ways of explaining current pulsations observed by us should also be discussed. As we know, current pulsations in nondegenerate semiconductors in strong, constant electric fields may be caused by the separation of the domains into high- and low-voltage, which is possible if their I-V characteristic is N shaped (e.g., Ref. 13). As applied to the present case, it would mean that the FM droplets remain stationary, and the field causes a separation of conduction electrons, which are heated by it, into the moving high- and low-voltage domains. The current pulse corresponds to a passing of a domain with increased conduction electron density through the cathode, after which the current drops sharply.

But the fact that the rectangular current pulse, which corresponds to the free carriers, retains constant height, and the peaks do not alternate with the valleys, is evidence of the nonexistence of the voltage domains in EuTe. Nevertheless, it should be noted that the N-shaped I-V characteristic is realistic for the degenerate AFM semiconductors with a phase separation. Although there is no evidence that the Gunn effect, which is related to the additional minimum in the conduction band, is possible in them, the N-shaped I-V characteristic may be a consequence of the Coulomb barrier for the recombination processes. In fact, such an I-V characteristic is typical of semiconductors with multiply charged impurity centers.[13] But from the point of view of semiconductor physics, each FM droplet plays the same part as the multiply charged center. Occupied by several electrons simultaneously, the FM droplet repels the conduction electron which goes from the conduction band to the droplet in the recombination process. Returning into this droplet, the electron should surmount this Coulomb repulsion. Heating by the external field favors the electron passing the barrier, which leads to a decrease in the conduction electron density. It is possible that this mechanism of the current pulsations can be realized in the other phase-separated degenerate semiconductors. It might also be possible to destroy the FM droplets by hot electrons, which would lead to the liberation of electrons trapped by droplets. This process is similar to the Auger ionization and for this reason should lead to an S-shaped I-V characteristic which manifests itself not in the current pulsations but in current pinching.[13] This effect might also be observed in systems under discussion.

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