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COOPERATIVE TRANSPORT PHENOMENA IN PHASE-SEPARATED DEGENERATE ANTIFERROMAGNETIC SEMICONDUCTORS

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

1. Introduction

The phenomenon of phase separation may realize in degenerate antiferromagnetic (AFM) semiconductors both displaying or not displaying the high-temperature superconductivity. It was first predicted and investigated theoretically still 20 years ago in papers [1], [2] (a detailed description of their results is given in [3], [4]). Then the phase separation was observed in nonsuperconducting EuTe [5], [6] and EuSe [7] (see an analysis of these experimental data in [3] and [4]). Recently phase separation was observed in superconducting systems based on AFM semiconductors (e.g. [8]).The problem of phase separation in such systems attracts much attention of theoreticians, too (e.g. [9], [10]).

In the present paper a new effect occuring in phase-separated AFM semiconductors EuTe is described: appearance of high current peaks over the Ohmic-type background in strong electric fields. These peaks form a periodic sequence with the period decreasing with increasing field strength. This phenomenon may be explained by cooperative motion of charged microregions of the ferromagnetic (FM) phase appearing 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. Specimens were grown by the chemical transport method in hermetically sealed Mo crucibles at 2000 C. Deficiency of Te was determined by temperature and exposition time. The sample length varied from 1,2 to 3 mm, their cross-section was 1 mm x 1 mm. As for their electric properties, at liquid nitrogen temperatures they behave like conventional n-type degenerate semiconductors, their conductivity being of the order of 100 $Ohm^{-1}cm^{-1}$ and the conduction electron densities of the order of 10^{19} cm^{-3}. (Similar properties exhibit crystals EuTe doped by I). But, on decrease in temperature, the conductivity of samples investigated drops sharply by several orders of magnitude (sometimes by the factor exceeding 10^6) reaching at 4.2 K values typical of nondegenerate semiconductors. A similar drop was observed earlier in [5], [6]. Magnetic measurements carried out in these papers confirm coexistence of AFM and FM phases in low-temperature region which means that this drop in the conductivity is a consequence of phase separation (see [3], [4]).

To avoid heating of the samples, impulse regime was used. Rectangular voltage pulses of a 10 μsec duration were applied to the samples at 4.2 K, the pulse frequence was from 1 to 10 Hertz. Most interesting are properties of samples with specific resistivity of the order of 10^3 Ohm cm at 4.2 K. In what follows, properties of a sample with the resistivity of $1.9 \cdot 10^3$ Ohm cm at 4.2 K will be presented in detail. Below the threshold value of the field strength which 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 to 20. The duration of increase and decrease of the current in the peak is found to be independent of the field applied in the field range investigated. This duration amounts to 0.7 μsec. On further increase in the voltage applied, 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. After this the third peak 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).

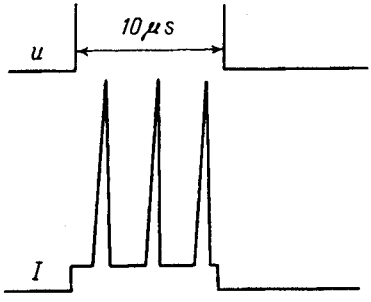

Рис.1

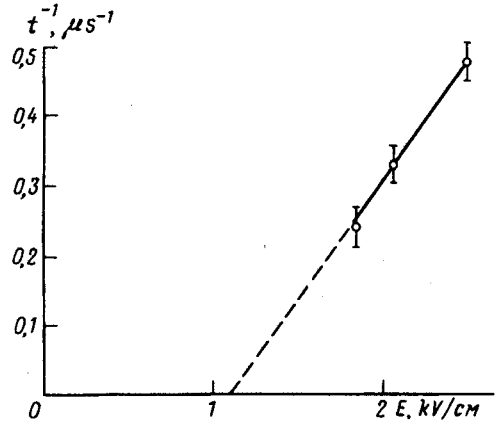

Рис.2

Fig. 1. Time dependence of voltage U and current I

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

An additional information may be obtained from the current-voltage characteristics of the sample recorded at the trailing edge of the current pulse with the strobe duration of 50 nanosec (Fig. 3). Below the threshold field at which the current peak appears for the first time, the height of the rectangular current pulse corresponds approximately to the Ohm's law (more precisely, the current dependence on voltage is superlinear). As seen from this Figure, after the current peak appearing 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, decreasing with the field. On the contrary, the peak width increases with the field. This is a result of the field dependence of the rate with which the peak moves over the current pulse.

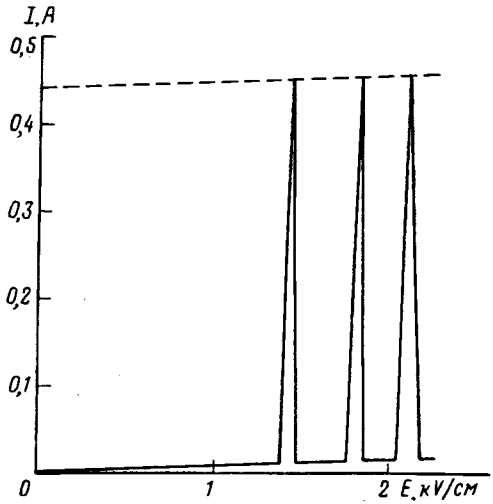

Fig. 3. Current I (A) versus aplied field E (kV/cm) with the strobe fixed at the trailing edge of the current pulse

Comparison of the results just presented with those for samples with resistivities ρ of $2 \cdot 10^3$ and $3.5 \cdot 10^3$ Ohm cm at 4.2 K does not show any ρ-dependence for the current pulse duration. 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 Neel point 9.6 K. They are absent at any temperatures for samples with resistivities at 4.2 K below than 10 Ohm cm and higher than 10^5 Ohm cm.

3. Theoretical interpretation

A natural way to explain these experimental results is to ascribe the rectangular current pulses to the charge transport by the conduction electrons of which the density is quite detectable at 4.2 K in the samples with current peaks (from 10^{13} to 10^{14} cm^{-3}). As for the current peaks, they may be explained by cooperative motion of the negatively charged FM microregions existing in the crystals EuTe.To make this point more clear it is instructive to give a brief survey of results [1],[2]

According to them, phase separation in degenerate AFM semiconductors is directly related to the ferron state in nondegenerate semiconductors first introduced in [11]: a charge carrier creates a FM region inside the AFM host and makes it stable by localization inside it. Virtually the ferron state corresponds to phase separation on a scale of a single carrier. In degenerate semiconductors phase separation is meant to be a cooperative ferron state in which the Coulomb interaction plays an important part. Thus, unlike phase separation at the a first-order phase transition, here separation of the crystal into two regions, one of them being FM and the other AFM, is impossible since the former would contain all the charge carriers and the latter be devoid of them at all. 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, its radius being of the order of several nm. With increasing carrier density n, the FM portion of the crystal converts from a multiply-connected region to the simply-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 whereas the latter case corresponds to a high-conducting state. Really, the set of FM droplets cannot move freely throughout the crystal since they are pinned by spatial fluctuations of electrostatic impurity potential inherent in degenerate semiconductors (certainly, they disturb also the periodic arrangement of the FM droplets). On the other hand, 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.

But in strong fields depinning of the FM droplets should occur, and they become able to move throughout the crystal as a united entity since they are connected with each other by Coulomb forces. Thus, a cooperative charge transport becomes possible like in systems with the charge density waves, though its regularities are quite different. Here they are determined by the fact that in the field the FM droplets retain their periodic arrangement. As 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 should vanish if the cathode borders with an insulating AFM layer situated between the droplet layers. Thus, as a function of time, the droplet current should display peaks between which it should vanish which agrees with the experimental data described above.

To make oneself sure that this explanation does not contadict our experimental data, one should compare theoretical values of volumes V_F for the ferromagnetic droplets with values of volume per droplet V_D estimated from experimental data. According to [2] the theoretical radius of droplets amounts to 1 - 3 nm, i.e., V_F is between $3 \cdot 10^{-20}$ and 10^{-19} cm^{-3}. The theoretical number of conduction electrons in each droplet N reaches several tens.

In order to estimate V_p, one should 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 corresponds to the cross-section area per droplet of from 10^{-13} to $5 \cdot 10^{-13}$ cm^2. Thus, the volume per particle V_p should be between $3 \cdot 10^{-20}$ and $4 \cdot 10^{-19}$ cm^3.

This volume includes both ferromagnetic droplet and its antiferromagnetic environment. In the geometry considered the ferromagnetic droplets 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 do not contradict this condition.

The specific feature of the cooperative transport under discussion is the fact that it realizes via hopping motion of droplets. One might think that the difference in the pinning potentials for different droplets should lead to incoherence in their motion which would manifest itself in spreading of the current peaks. But one should keep in mind that the interaction between droplets may help surmount the pinning potentials by all the droplets simultaneously: the system of the FM droplets should respond to a certain average pinning potential. This prevents current peaks from spreading. Moreover, one may prove that the correlations between the droplets may increase their probability of hopping.

To illustrate the latter statement, a simplified model will be treated: a pair of particles modeling the FM droplets is initially located on a pair of neighboring sites A and B. Then the particles go over from the site A to the site C and

from the site B to the site D, respectively, the sites C and D being neighbors, too. The Hamiltonian of the system is of the form:

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

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

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

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

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

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

Then, considering H_0 as the zeroth-order Hamiltonian and H_1 as the perturbation, one obtains the following expressions for the probability $W_{AB,CD}$ for a pair of particles to go over jointly from the atoms A, B to atoms C, D at time t large enough:

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

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

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 taking into account the interaction of FM droplets with phonons (like in [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 should be peaked at $E = 0$.

As one sees from comparison of (6) and (8), the intersite hopping of a particle not interacting with another particle is forbidden at $K \neq 0$. But interaction with another particle may make their joint transition allowed if $K = U$. One should note that the transition probability $W_{AB,CD}$ for uncorrelated particles (6) is the product of transition probabilities for separate 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 consideration presented above. This effect is independent of the sign of U. But one should keep in mind that such a motion is possible if only the interparticle correlations are not too strong. In fact, even if one replaces the δ-functions by $\rho(E)$ in (8), the probability should vanish like $1/U^2$ with increasing U.

One should discuss also other possible ways of explaining current pulsations observed by us. As well known, current pulsations in nondegenerate semiconductors in constant strong electric fields may be caused by the separation into high- and low-voltage domains which is possible if their IVC is N-shaped (e.g., [13]). As

applied to the present case, it would mean that the FM droplets remain stationary, and the field causes separation of conduction electrons, heated by it, into moving high- and low-voltage domains. The current pulse would correspond passing of a domain with increased conduction electron density through the cathode, after which the current should drop sharply.

But the fact that the rectangular current pulse corresponding the free carriers retains the constant height, and the peaks do not alternate with valleys, evidences nonexistence of voltage domains in EuTe. Nevertheless, one should note that the N-shaped IVC is quite realistic for degenerate AFM semiconductors with phase separation. Though there are no evidences that the Gunn effect related to an additional minimum in the conduction band is possible in them, the N-shaped IVC may be a consequence of the Coulomb barrier for the recombination processes. In fact, such an ICV 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 muliply-charged center. Being occupied by several electrons simultaneously, the FM droplet repulses a conduction electron which is going over from the conduction band to the droplet in the recombination process. For returning into this droplet, the electron should surmount this Coulomb repulsion. The heating by the external field favors electron passing the barrier which leads to drop in the conduction electron density. Possibly, this mechanism of current pulsations may realize in other phase-separated degenerate semiconductors.

One might also think of destroying FM droplets by hot electrons leading to liberation of electrons trapped by droplets. This process is similar to the Auger ionisation and for this reason should lead to a S-shaped IVC which manifests itself not in current pulsations but to current pinching [13]. This effect might be observed in systems under discussion, too.

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