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# Tunneling Magnetoresistance in a $Eu_{0.7}Pb_{0.3}MnO_3$ (Single Crystal)–Fe (Film) Structure

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**Abstract**—We have studied the magnetoresistive properties of a structure comprising single crystal manganite $Eu_{0.7}Pb_{0.3}MnO_3$ covered with an epitaxial iron film. At temperatures below $T_C$ of the manganite crystal, the structure exhibits positive magnetoresistance. The behavior of the resistance as a function of the magnetic field is characteristic of a tunneling junction with ferromagnetic electrodes separated by a thin insulating film. The observed effect is related to the formation of a transition layer at the manganite–Fe interface, which is depleted of oxygen and possesses dielectric properties. The sensitivity of the resistance with respect to the magnetic field is determined both by the negative magnetoresistance of the manganite crystal and by the tunneling contribution to the conductivity, whereby the tunneling current depends on the mutual orientation of magnetic moments of the electrodes ($Eu_{0.7}Pb_{0.3}MnO_3$ crystal and Fe film). © *2003 MAIK "Nauka/Interperiodica".*

Investigation into spin-dependent transport via heterostructures containing magnetically active layers is among currently important and highly promising directions in the physics of magnetic phenomena. These hopes are related to the large application potential of such structures [1]. Indeed, such structures, retaining all advantages of the traditional semiconductor (nonmagnetic) low-dimensional systems, acquire an additional channel of control via magnetic field [2]. This possibility greatly expands the functional properties of microelectronic devices. One of the related problems encountered in this way is the search for new materials providing for a high spin polarization of electrons emitted into a magnetic structure. A promising solution is offered by ferromagnetic semiconductors based on magnesium oxides with a perovskite structure, known as manganites. These compounds are characterized by a high degree of spin polarization of the intrinsic carriers, reaching nearly 100% [3].

Below, we report on the results of investigations of the magnetoresistive properties of a structure comprising single crystal manganite covered with an epitaxial iron film ([M/Fe]). The single crystals of $Eu_{0.7}Pb_{0.3}MnO_3$ manganite were grown by spontaneous crystallization from a solution melt. The samples exhibited a transition to the ferromagnetic state with a metal conductivity at $T_C$ = 210 K. The substrates cut from single crystal ingots had the shape of 3 × 2 × 0.1 mm plates, in which the large plane coincided with one of the principal crystal planes. An iron film with a thickness of 250 Å was deposited by molecular beam epitaxy on a commercial setup of the Angara type. Copper films with thicknesses up to ~500 Å deposited in the same technological cycle onto the outer iron film surface and onto the bottom substrate surface were used as electrodes during electric measurements on the [M/Fe] structures. The experimental geometry is outlined in the inset to Fig. 1.

Figure 1a shows a plot of the resistance $R(H)$ and magnetoresistance $(MR)\Delta R/R_0 = (R(H) - R(0))/R(0)$ as functions of the applied magnetic field for the [M/Fe] structures studied at $T$ = 80 K. For comparison, Fig. 1b presents an analogous curve for the substrate (single crystal manganite). As can be seen, the $R(H)$ curve for the [M/Fe] sample exhibits a characteristic maximum representing the region of positive MR. As is well known, manganites proper exhibit a negative MR effect, which is clearly observed for the single crystal of $Eu_{0.7}Pb_{0.3}MnO_3$. A comparative analysis showed that, in magnetic fields above $H$ ~ 11 kOe, the change in the sample resistance is determined entirely by the MR properties of the substrate. Since the resistance of the [M/Fe] structure is about three times that of the substrate, it is natural to suggest that the features of $R(H)$ of the structure are related to the formation of a transition layer between the iron film and the manganite substrate.

A peak corresponding to the positive MR region on the $R(H)$ curve begins to appear in weak magnetic fields at $T < T_C$. As the temperature decreases, the maximum $R_m = R(H_m)$ shifts toward greater magnetic fields ($H_m$ is the field strength corresponding to the maximum value of $R(H)$) and the magnitude of the MR effect ($\Delta R_m = R_m - R_0$) increases. On the whole, the character of the

temperature variation of $\Delta R_m$ and $H_m$ is correlated with the behavior of magnetization of the manganite crystal. It is important to note that the region corresponding to positive MR values appears only in the interval of temperatures at which the manganite crystal possesses a spontaneous magnetic moment.

Another important feature is that the magnitude of the MR effect depends on the bias voltage $V$ applied to a sample during the resistance measurements. This behavior is characteristic of the tunneling transitions between electrodes of a ferromagnetic material separated by a thin dielectric layer ($[FM_I/I/FM_{II}]$ structures). The presence of the $R_m$ peak is related to the spin valve effect [3], the tunneling current being dependent on the magnetic moments of electrodes. The contact resistance is minimal when these moments ($\mathbf{M}_I$ and $\mathbf{M}_{II}$) are parallel to each other, and the resistance is maximal when the moments are antiparallel. In our case, the role of the magnetically active electrodes is performed by the iron film and manganite crystal (for $T < T_C$). Additional evidence for the proposed mechanism of the tunneling MR in the structure studied is provided by the current–voltage characteristic (Fig. 2) possessing a shape typical of the tunneling junctions [4]. In contrast, the current–voltage characteristic of the substrate (single crystal manganite) is linear.

Let us consider the possible mechanism of the formation of a transition layer between a manganite crystal and an epitaxial iron film. As is well known, manganites are characterized by a weak binding of oxygen to the lattice and a relatively easy diffusion of this component. On the other hand, manganites are extremely sensitive to deviations from the crystal stoichiometry in oxygen [5]. For some compositions, a decrease in the oxygen content resulted in the sample (exhibiting ferromagnetic/conducting properties in the stoichiometric state) acquiring dielectric properties with an antiferromagnetic type of ordering [6]. Thus, it seems that the stoichiometry with respect to oxygen is a decisive factor determining the properties of near- surface layers in manganites, including the transition layers between the crystal substrate and a film.

In our case, the situation can be as follows. The interface between the crystal and the film features mutual diffusion that results in the formation of a thin layer depleted of oxygen as compared to the stoichiometry. This layer, possessing dielectric properties, plays the role of a potential barrier for electrons. Using the classical expression of Simmons [4] for the tunneling current via a potential barrier, we calculated a current–voltage characteristic approximating the curves experimentally measured for the [M/Fe] structures. From this we estimated the height $U_0$ and width $d$ of the aforementioned potential barrier: $U_0 \approx 29.5$ meV and $d \approx 20$ Å. Obviously, these estimates are quite rough and can be considered as a lower boundary for the real values of $U_0$ and $d$, since the tunneling mechanism of conductivity is probably not dominating at high temperatures ($T > 80$ K) and the over-barrier transport of charge carriers (thermoelectron emission) has to be taken into consideration as well. In addition, the interfacial region is likely to be inhomogeneous [7] and may contain microscopic highly conducting inclusions percolating through the barrier.

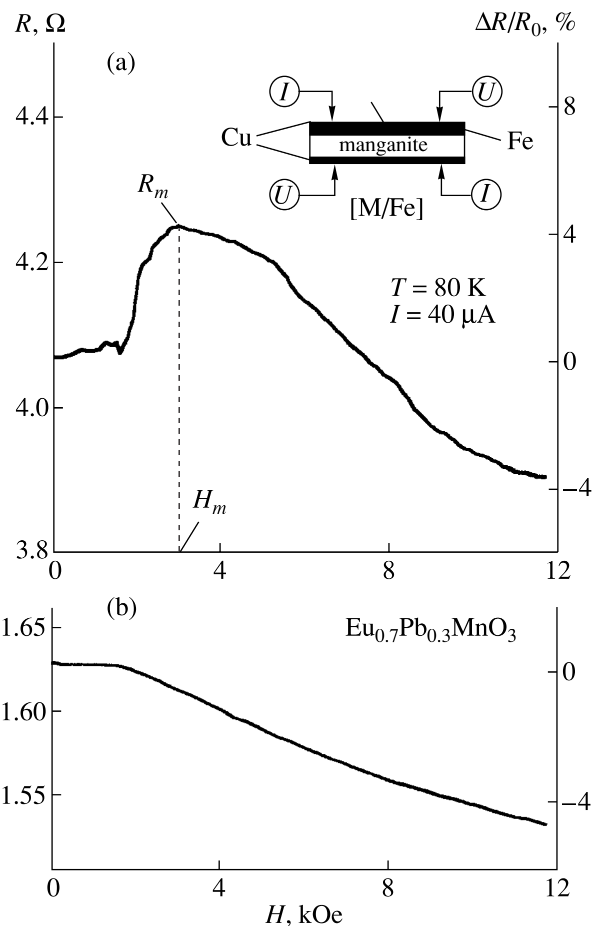


**Fig. 1.** Plots of the resistance $R(H)$ and magnetoresistance $\Delta R/R_0 = (R(H) - R(0))R(0)$ versus the applied magnetic field at $T = 80$ K for (a) the [M/Fe] structure studied and (b) the substrate ($Eu_{0.7}Pb_{0.3}MnO_3$ single crystal). The inset shows a schematic diagram of the experimental arrangement.

The spin valve effects are traditionally observed and characterized using the structures in which ferromagnetic films are separated by dielectric spacers, so that the electrodes are magnetically uncoupled. A difference in the behavior of magnetic moments $\mathbf{M}_I$ and $\mathbf{M}_{II}$ as the functions of $H$ and, hence, a change in the tunneling current via the junction can be realized by selecting the materials for $FM_I$ and $FM_{II}$ possessing different coercive fields ($H_{CI}$ and $H_{CII}$, respectively). In this case, the $R(H)$ curve exhibits a considerable hysteresis. No such hysteresis was observed in the [M/Fe] structures studied in our experiments. Another distinctive feature is that the magnetic field strength ($H_m$) corresponding to the maximum resistance $R_m$ has a large value ($H_m$ =

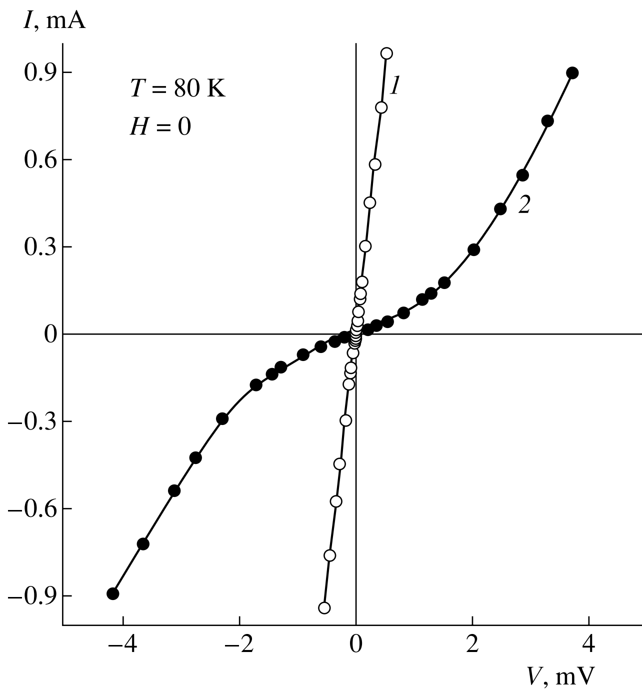


**Fig. 2.** Current–voltage characteristics of (*1*) a $Eu_{0.7}Pb_{0.3}MnO_3$ single crystal and (*2*) a [M/Fe] structure at $T = 80$ K.

3.3 kOe), which is significantly greater than the analogous $H_C$ values for $Eu_{0.7}Pb_{0.3}MnO_3$ crystals and iron films taken separately.

This behavior can be rationalized by assuming that the dielectric transition layer possesses magnetic properties and mediates an exchange interaction between the manganite crystal and the iron film, this interaction possessing an antiferromagnetic character (as was indicated above). The equilibrium angle θ between the magnetic moments $\mathbf{M}_I$ and $\mathbf{M}_{II}$ is determined by the condition of the minimum free energy density:

$$F(\theta) = F_{EX} + E_Z + E_D + E_A,$$

where $E_{EX}$ is the energy of the exchange interaction between electrodes, $E_Z$ is the Zeeman energy in the applied field, $E_D$ is the energy of a demagnetization field, and $E_A$ is the energy of the magnetic anisotropy. A possible scenario for the tunneling contribution to $R(H)$ is as follows. For $H = 0$, both the manganite and the iron film occur in a multidomain state with all relative orientations of $\mathbf{M}_I$ and $\mathbf{M}_{II}$ in the plane being equiprobable. As the field strength $H$ increases, the magnetization proceeds and a saturation magnetization of the crystal and film is attained at $H \approx H_m$. The antiferromagnetic exchange via the near-surface layer results in an approximately antiparallel orientation of $\mathbf{M}_I$ and $\mathbf{M}_{II}$ and the structure is characterized by a maximum value of $R(H)$. As the field $H$ increases further, competition (mostly between $E_{EX}$ and $E_Z$ contributions to $F(\theta)$) takes place, the equilibrium angle θ decreases, and $R(H)$ drops. For $H \sim 11$ kOe, the moments $\mathbf{M}_I$ and $\mathbf{M}_{II}$ become parallel; at still higher fields, the MR of the structure is dominated by the contribution of the substrate, which is not related to the tunneling mechanism.

The proposed scenario qualitatively explains the observed behavior of MR in the [M/Fe] structure studied. The situation will be elucidated in greater detail by investigations of the behavior of the magnetizations of the manganite crystal and the deposited iron film.

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