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Giant Magnetoresistance in the Polymer-Ferromagnetic System

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A new magnetoresistance phenomenon is observed in a magnetic—polymer—non-magnetic metal multilayer structure. For the first time, giant magnetoresistance is obtained on the ferromagnetic- poly(arylenephthalide) boundary and can be switched on and off by varying the external magnetic field. We present the results of researches of the ferromagnet—polymer—non-magnetic metal system that has all above-mentioned properties at room temperature. The magnetoresistance is extremely large, so the working cell resistance changes at least by 10^4 times. The current-voltage characteristics of the magnetic—polymer—Cu structure with giant magnetoresistance 'on' confirm the influence of the magnetic field on the system.

Keywords: fermi level; giant magnetoresistance; polymer film

PACS: 75.47.De

The electron spin direction is surely a parameter, whose control is of great interest. Controlling the spin direction, one will be able create a new generation of electronic devices in the future.

The research of semiconductors [1,2] showed that these materials can change magnetoresistance in the external magnetic field, correlating with the spin-dependent surface contact resistance [3,4]. In 2000, P. LeClair and others [5] discovered that the tunnel magnetoresistance in an Al_2O_3 film between two ferromagnetic electrodes is

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proportional to the product of the density of spin polarized states in the left and right electrodes. It was marked also that a change of the chemical potential in a magnetic field can indicate the type of a magnetic state of the substance [6]. In [7,8], the authors suggested to use the ferromagnet—semiconductor—ferromagnet system for the creation of spin-polarized transistors.

The question of the existence of high conductivity in low-dimensional systems was also discussed in [9], as well as the coexistence of high conductivity and ferromagnetism [10]. Today the organic polymer materials are considered to be the most perspective low-dimensional systems. It was suggested to use the existence of electron traps in a polymer layer for the current amplification [11] and for the creation of memory elements [12].

Nevertheless the spin-dependent magnetoresistance in the previously researched systems either was detected under extremely low temperatures [13–15] or was not great [16,17]. The theoretical research into the spin-charge manipulation abilities [18] assumes complex experimental devices for practical creation. But, for obtaining the devices working on the base of spin polarization, one needs the simple structures which must possess high magnetoresistance, ferromagnetism, and high room-temperature conductance simultaneously. Moreover, one should be able to change these properties in the desired direction. That's why some unusual polymer capabilities [19] took our attention. We also had known about the indication of the electrode's magnetic phase transition for such systems [20] with the help of a polymer layer.

Now a series of investigations of the electronic switch of the conductivity of metal—polymer—metal structures has been accomplished [21]. It was shown that these systems had been extraordinary sensitive for external influences such as pressure, temperature, and the phase composition of electrodes. So the subject of researches today is how the metal—polymer—metal cell indicates the change of a magnetic state of ferromagnetic electrodes. At first, we take only one electrode to be ferromagnetic in order to see the effect clearly.

The experimental device includes the source of stabilized voltage, digital voltmeter, two-coordinate recorder, magnetic field meter, standard laboratory electromagnet, and ballast resistor for the circuit current limitation.

The sample was prepared as a multilayer structure of the magnetic—polymer film—non-magnetic metal type. A plate of ~ 1.5 mm in thickness was used as the magnetic metal. It was made of polycrystalline Ni. Ni was chosen as the spin polarizer of injected electrons, as it was a strong zone magnetic, and, as was estimated in

[22], the split of the electron states near the Fermi surface for the electrons with opposite spin directions in this material might be of very large value $\Delta \sim 1\,\text{eV}$. The second metallic electrode was prepared by vacuum evaporation of Cu. To minimize the influence of the magnetostrictive effect arising in a magnetic field, the ferromagnetic plate was made as a plate, composed of Ni contacted with the polymer film and the Fe plate having the same area and thickness. The Fe plate was attached to the Ni plate on the side opposite to the polymer. The magnetic field was applied perpendicularly to the polymer surface.

The polymer film was made of a polymer of the poly(arylenphthalide)s class. Poly(3,3′-phtalydiliden-4,4′-biphenylen) has molecular mass of $(50–80)\cdot 10^3$. The absence of the electron-hole symmetry in these polymers is an important particularity of their electron structure. Quantum-chemical calculations showed that the electron states forming the highest occupied molecular orbital (HOMO) are localized on the molecule backbone, and the electron states corresponding to the lowest unoccupied molecular orbital (LUMO) are localized on the side fragment of the molecule [23]. In connection with this fact, one can propose the electron-hole excitation absence, and so the collective spin-wave mode quenching is small in this polymer.

Previously it was shown that these polymers possess the unique electronic properties – one can induce a phase transition of the dielectric-metal type affecting the metal-polymer-metal type structure by extremely small influences, such as electric field, pressure etc. [24].

The dependence of the polymer film resistance on the pressure is presented in Figure 1. The initial resistance of the sample, R, was more than 10^8 ohm at $P = 0\,\text{Pa}$. The film resistance begins to decrease with reaching the critical pressure which was $P_{\text{CR}} \sim 5\cdot 10^4$ Pa for this polymer sample. The decrease in the resistance takes place in a narrow pressure interval. The resistance reaches $R_{\min} \sim 0.1$ ohm and stays constant with the next pressure increase. The cause of such resistance behavior has been discussed earlier in some works [23,24]. It is connected with the existence of a narrow conduction band in the middle of the polymer energy gap [15].

In the present work, the initial resistance (while $B = 0$) of the multilayer structure was set by applying a pressure near the high-conductivity transition point. The maximum pressure did not exceed 0.5 MPa.

Polymer films of $\sim 800 \pm 50\,\text{nm}$ in thickness were prepared by the spin casting method from a 5% polymer solution in cyclohexanone. The film thickness was estimated with a microinterferometer MII-4.

The dependence of the current passing through a sample on the magnetic field is presented in Figure 2.

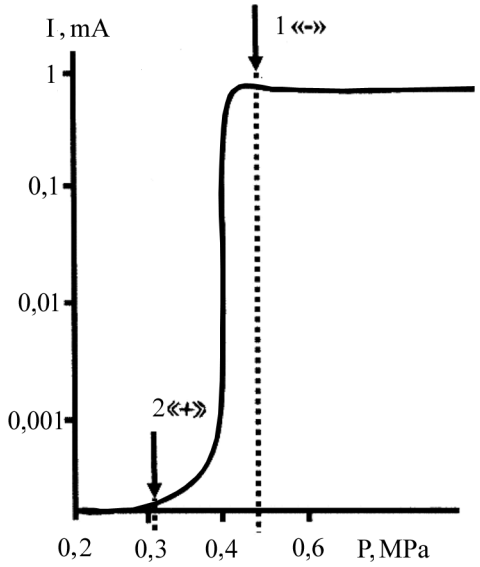

FIGURE 1 Choice of the initial state of a sample. The current flowing through the ferromagnet—polymer—metal system vs. uniaxial pressure. Arrows show the areas of pressure chosen for providing the initial resistant state required. Signs (+) and (−) correspond to the sign of the GMR effect obtained.

The initial sample state is a low-resistance one at $B = 0$. With increase in the magnetic field, the conductance of the sample was practically constant up to $B \sim 130\,\text{mT}$. Near this point, the current changes of the fluctuation character were registered in the measuring circuit. The fluctuation amplitude increased with H. The current decrease took place as the magnetic field reached the value of $B = 160\,\text{mT}$, and the sample nearly switched into a high-resistance state. The relative current changes were of 4–5 orders of magnitude. The next increase in the magnetic field did not lead to the current change.

While the magnetic field decreases, the sample experiences the back transition into the initial low-resistance state, but at a less value of the magnetic field, $\sim 148\,\text{mT}$. That is, the hysteresis is observed. The multiple repeating of the measuring cycle reproduced the magnetic field threshold values for the resistance changes with great exactness. This effect was reproduced many times with a lot of samples.

The analysis of the obtained results and the comparison with the data published for analogous metal/polymer structures show that

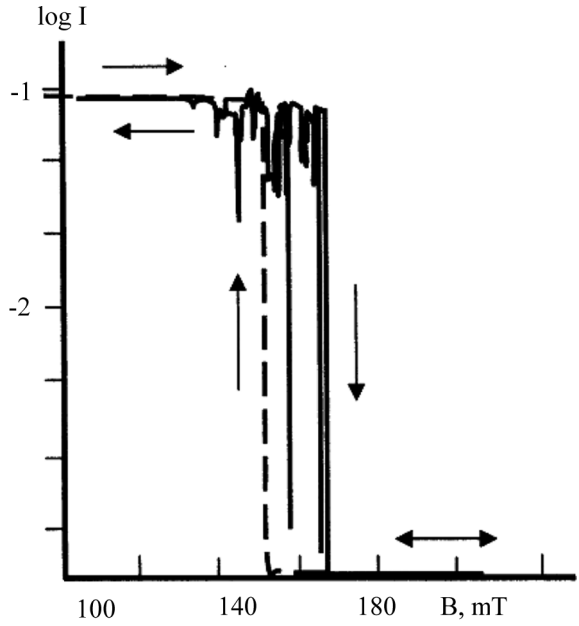

FIGURE 2 Current flowing through the Ni—polymer—Cu structure vs. magnetic field value. Arrows point the magnetic field change direction. The solid line corresponds to the increase in the external magnetic field, and the dotted line corresponds to its decrease. The voltage applied to the experimental structure was 1 V.

there are several remarkable particularities in the considered case. We underline some of them:

1. Resistance change takes place at respectively high room temperature.
2. Resistance change is unusually large. The largest values for analogous configurations using an organic material as the transport layer did not exceed several percents [24]. In our case, the maximal registered resistance change was of 5 orders of magnitude.

We propose that the nature of such large resistance change caused by the magnetic field is different as compared to that traditionally considered for such systems. Apparently, the magnetization increase of the Ni electrode leads to the increase of the spin-polarized part of electrons on the Fermi level. The measured magnetization curve for Ni and Ni + Fe samples shows that the saturation area was not

reached in the experiments performed. It is known that the Fermi-level positions for the spin-up and spin-down electrons in magnetic materials are different. The initial state of the Ni—polymer—Cu sample (at $B = 0$) was low-resistance. According to work [21], in this state, the narrow partially occupied subband is formed in the polymer. It is placed in the middle of the polymer energy gap. The effective charge transport is possible in this case in the metal-polymer-metal system, if the position of the injecting level of the metal (the Fermi level) coincides with the position of the narrow subband in the polymer band gap. The current flowing through such a border depends exponentially on the energy distance between the Fermi level in the metal and the narrow subband in the polymer. Apparently, the Fermi level position changes in the magnetic material under the action of an external magnetic field, which appears to be the main reason for such abrupt change of the resistance in the Fe/Ni-polymer-Cu system.

To confirm the assumption about such an influence of the magnetic field on the charge transfer in the experimental structure "magnetic-polymer–non-magnetic metal," the current-voltage characteristics were investigated with different values of the magnetic induction (Fig. 3). It was established that GMR strongly depends on the current direction.

If the explanation proposed is correct, then the conductivity change effect in the ferromagnet-polymer system must take place also when the initial state is high-resistance. High resistance can mean that the Fermi level and the narrow subband related to the coherent charge

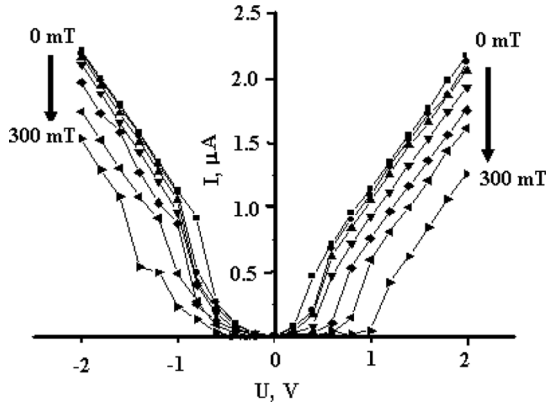

FIGURE 3 Current-voltage characteristics of the Fe-Ni—polymer—Cu structure with different values of the external magnetic field (0–300 mT). Arrows showed the direction of the magnetic field increase by a step of 50 mT.

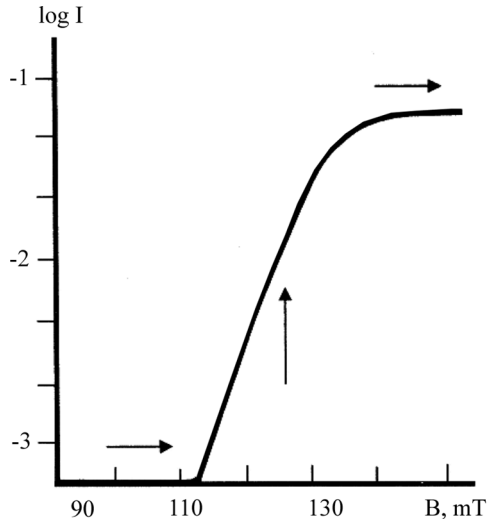

FIGURE 4 Current flowing through the Ni—polymer—Cu structure vs. magnetic field value. The initial state is high-resistance. After the threshold field, we get the low-resistance state. The voltage applied to experimental structure was 1 V.

transport are not isoenergetic. The external magnetic field is capable to compensate the energy difference. In this case, one will observe a considerable decrease of the resistance in the system under study.

The result of such an experiment is presented in Figure 4. The initial sample resistance was set at a small uniaxial pressure. The pressure value was chosen near the switching threshold, but a little bit less. In Figure 1, it is marked with arrow 2. The sample resistance in this state was of 10^5 kohm. When the magnetic field reached the value of $B = 110$ mT, the abrupt resistance drop took place. The sample resistance decreased down to less than 100 ohm and stayed at this level with the further growth of the magnetic field.

Thus, we have discovered the effect of electronic switching induced by a magnetic field in the ferromagnet–electroactive polymer system.

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