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# Magnetization reversal of ferromagnetic nanoparticles induced by a stream of polarized electrons



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### ABSTRACT

The remagnetization of ferromagnetic Fe<sub>3</sub>O<sub>4</sub> nanoparticles of several thousand cubic nanometers by spin-polarized current is investigated. For this purpose, magnetite nanoparticles are synthesized and deposited on a conductive nonmagnetic substrate. The remagnetization is conducted in high-vacuum scanning tunneling microscope (STM). The STM tip from magnetized iron wire constitutes one electrode while the ferromagnetic nanoparticle on the graphite surface represents the second electrode. The measured threshold value of remagnetization current ( $I_{thresh}=9$  nA) is the lowest value of current at which remagnetization occurs. The change in nanoparticle magnetization is detected by the effect of giant magnetic resistance, specifically, the dependence of the weak polarized current ( $I < I_{thresh}$ ) on the mutual directions of magnetization of the electrodes. The results indicate essential difference with available literature data on the influence of polarized current on magnetic moment of small ferromagnetic nanoclusters. The peculiarities of size dependence of the observed effects are explained.

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# 1. Introduction

The dependence of current between two ferromagnetic electrodes separated by a thin insulating layer or a nonmagnetic metal on the mutual orientation of the magnetizations in the electrodes, i.e. the effect of giant magnetic resistance (GMR), has been known since 1988 [1,2]. This effect is widely used for reading in magnetic information recording systems. In 1996, Slonczewski [3] proposed a possible theoretical solution which was somehow the inverse problem of remagnetization of magnetic domain by polarized current, i.e. by flow of electrons with a preferred spin direction. The study considered the current as a ballistic flight of electrons from one ferromagnetic electrode to another.It allowed for the interaction of the spin magnetic moment of current electrons, which is parallel to the magnetic moment of the ferromagnetic cathode, with the magnetic moment of the ferromagnetic anode. Such an interaction may result in a change in the direction of anode magnetization. At about the same time, Berger discussed the possibility of a magnon excitation in a ferromagnetic material by the polarized current [4]. The study allowed for the excitation of a magnon that leads to a reduction of domain magnetization on one magnetic moment of the electron. The possibility of domain

remagnetization by the current in a massive ferromagnetic was also investigated theoretically [5]. The modified Landau–Lifshitz kinetic equation was applied for characterization of the domain remagnetization. This modified equation took into account the interaction of the magnetic moment of polarized current with the domain magnetic moment. Such an interaction may cause a change in the stationary domain magnetization.

The possibility of massive magnetic domain remagnetization by polarized current has been demonstrated experimentally [6–8]. The observed transverse dimension was on the order of microns, and the thickness was  $\sim 10$  nm. Thus polarized current was introduced through a point contact, causing the rotation of magnetization in the (Co–Cu)N multilayer sandwiches [6]. A switch in the orientation of the magnetic moments in (Co–Cu–Co) sandwich structure was also observed for perpendicular electric currents throughout the domain plane [7]. In a subsequent study [8] the change of magnetic domain polarization was attributed to the diffusion current of polarized electrons through the non-magnetic conductor from another ferromagnetic.

The standard procedure often used to study and change nanoparticle magnetization is the scanning probe microscope, typically the Atomic Force Microscope (AFM) or the Scanning Tunneling Microscope (STM), with its own magnetization. For example, the remagnetization was studied of an ordered array of nanoparticles with a diameter 5–20 nm and a height of about

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100 nm consisting of cobalt and iron–chromium alloy, utilizing the field of the permanent magnet on the probe of the atomic force microscope [9]. This technique was demonstrated to allow both the diagnosis of magnetization vectors as well as study their effect readily. A magnetic STM chromium tip indicated double chains of atoms, consisting of a few tens of atoms located on the surface of Ir (001) with ( $5 \times 1$ ) reconfiguration and its characteristic magnetization vector [10]. This result established the change of chain magnetization.

There is an alternative approach to study magnetic nanoparticles and the effect on their magnetic characteristics, based on irradiation of a sample by the current of spin-polarized electrons. This technique was used to investigate the magnetic properties of nanostructured coatings consisting of iron nanoparticles deposited on a monocrystalline surface of W(001) [11]. The nanoparticles used were in the 20–40 nm size range and height of less than 1 nm. In particular, the study provided the morphological and electronic structures, their dependence on spatial distribution on the substrate and the magnetization vector. In the latter case, the spin-polarized tunneling current was obtained with a tungsten tip covered by a layer of iron.

Further development of these techniques has led to the method of spin-polarized scanning tunneling microscopy (SP-STM), which allows investigation and modification of the magnetic moments of nanostructures. Thus, at sufficiently high densities of the tunneling current, the island consisting of 100 atoms of iron [12] changes its magnetization. In addition, it has been shown that the SP-STM allows the study of reversible magnetization of the island with lateral diameter of about 2 nm, consisting of 40 atoms of iron. In this case the threshold value of the tunneling current is about 1  $\mu$ A [13]. A continuation of the above studies also considered the effects of magnetic moment rotation of superparamagnetic Co nanoislands due to the tunneling current [14].

Based on the aforementioned studies [12–14] the proposed mechanism of remagnetization of ferromagnetic nanoclusters in the STM includes the heating by the current. Even a small amount of heat, on the order of 1 K (corresponding to 600 nA current), at the sample temperature of 30 K affected the stability of magnetization direction of the small (on the order of several tens of atoms) ferromagnetic island [14]. Thus the typical operating values of spin-polarized tunneling current and the pulse duration were microamperes and hundreds of milliseconds respectively [13]. These values were obtained for particles of about 2 nm size and less than 1 nm in height.

It should be noted that the theory proposed previously [3–5] is based on the following assumption: the total spin momentum of the electrons directed along the magnetization of the ferromagnetic cathode is stored as an integer within the domain. The remagnetization process is described as the result of interaction of two macroscopic magnetic moments – the domain moment and the current moment, i.e. it is assumed that the spin moments of current electrons remain coherent inside the anode.

An alternative approach to the remagnetization description of individual nanoparticles by the current was presented in a previous study [15]. The study considered the remagnetization of ferromagnetic nanoparticles on a non-ferromagnetic substrate in a scanning tunneling microscope (STM) comprised of ferromagnetic tip. The approach is based on the kinetic equations for the number of electron spins directed along and against the direction of magnetization. These numbers are distinct from the equilibrium values when the current flows through a magnetic nanoparticle. It was found that the nanoparticle remagnetization is possible only if the current is greater than a certain critical current  $I_{cr}$ .

The value of  $I_{cr}$  depends on the size of nanoparticle and the degree of tip and nanoparticle magnetization. And the time of remagnetization tends to infinity, when the current is close to  $I_{cr}$ .

The estimation of  $I_{cr}$  for ferromagnetic nanoparticle of ~10 nm size gives a value of about 10 nA [15].

In contrast to previous experiments investigating remagnetization of small ferromagnetic nanoclusters [12–14], the present study is concerned with large nanoparticles ( $\sim 15 \times$  $50 \times 50$  nm<sup>3</sup>). A scanning tunneling microscope is employed in which one electrode is the ferromagnetic tip and the other electrode is ferromagnetic nanoparticles on the graphite substrate. The remagnetization of nanoparticles by the electron flow from a ferromagnetic STM tip is investigated. In the first stage the tip is the anode and the magnetization of the tip and nanoparticles coincide. In a subsequent second stage, the process is reversed. With the change of electrical polarity, when the tip represents the cathode, the nanoparticle magnetization is altered as a result of the current flow, and magnetization of the nanoparticle and magnetization of the tip are directed to one side. In addition, the critical currents of remagnetization are measured.

# 2. Theoretical outlook

According to the theory developed in previous studies [3,4], the total spin angular momentum of electron flow directed along the magnetization of the ferromagnetic cathode is preserved within the domain (i.e. the anode), with a different magnetization direction. In other words, the spin wave functions of the electrons in the flow should also conserve their phases in the domain.

However, only the projections of electron spins along or against the direction of the domain magnetization are conserved in the current. The transverse components of the spins are not conserved due to collisions with other electrons of a ferromagnetic (collision frequency is  $10^{13} \div 10^{14} \text{ s}^{-1}$ ). The spins are oscillating with different phases due to incoherent scattering of the different electrons of the current on the domain electrons. Therefore, the resulting transverse component of the spin can be considered equal to zero. The concentrations of electrons with spins along and against domain magnetization as the current passes through nanoparticles are different from their equilibrium values.

The kinetic equations can be written for these concentrations which depend on the input and output flows of electrons with the different spin directions. Assume for simplicity that the magnetization directions in a ferromagnetic electrode (STM tip) and nanoparticles are collinear, i.e. they are directed along or against each other. Let us determine the electron flows from the ferromagnetic electrode to the cathode in the nanoparticle with the directions of spin along ( $I_{+}^{ent}$ ) and against ( $I_{-}^{ent}$ ) spin polarization in the nanoparticle. At room temperature, the characteristic relaxation time of the energy and moment of an electron which tunnels from one electrode to another and a hole that remains in the first electrode is about  $10^{-14}$ – $10^{-13}$  s.

Tunneling currents which will be considered here are  $I \le 10^{-7}$  A. The average time duration between the passages of the electrons is  $\tau_{inter} \ge 10^{-12}$  s. Therefore, the tunneling current flows between the electrodes which are in the equilibrium state.

As was shown in a previous paper [15], if the magnetization directions of the tip and nanoparticles are opposite, then

$$I_{+}^{ent} = I \frac{\alpha}{\alpha + \alpha_p}, \quad I_{-}^{ent} = I \frac{\alpha_p}{\alpha + \alpha_p}$$
(1)

Here, we have introduced the notations  $\alpha = \rho^{-}/\rho^{+}$  and  $\alpha_{p} = \rho_{p}^{-}/\rho_{p}^{+}$ , where  $\rho^{-}$ ,  $\rho^{+}$  are the electron densities of states at the Fermi surface with spins against and along polarization in the electrode (STM tip) and  $\rho_{p}^{-}$ ,  $\rho_{p}^{+}$  are the same quantities in the nanoparticle. Assuming the dispersion law of *p*-electrons in ferromagnetic nanoparticles  $\varepsilon = p^{2}/2 m^{*}$  (here  $m^{*}$  is effective electron

mass) and using the expression for density of states of a Fermi gas, we obtain the following expression:

$$\rho_p^{+,-} = \frac{3^{1/3}}{\pi^{4/3}} m^* (n_p^{+,-})^{1/3}$$
Hence,
$$\alpha_p = \left(\frac{1-x}{1+x}\right)^{1/3}$$
(2)

where  $x=(N_+-N_-)/N$ , and  $N_+$  and  $N_-$  are respectively the numbers of electrons with spins directed along and against the direction of initial polarization. The expressions for outgoing currents from the nanoparticle to non-ferromagnetic electrodes assume the form (see also the review on spintronics [16]):

$$I_{+}^{ex} = I/(1 + \alpha_p), \quad I_{-}^{ex} = I\alpha_p/(1 + \alpha_p)$$
 (3)

Knowing the expressions for the incoming and outgoing polarized currents it is possible to calculate the kinetics of remagnetization. Let us find the change in the quantity of oppositely polarized electrons in the nanoparticles due to the current. It is obvious that the currents  $I_{+,-}^{ent}$  will increase these values, and the currents  $I_{+-}^{ex}$  will reduce them. In addition, it is necessary to allow for the relaxation of polarization in the ferromagnetic to the equilibrium value. Such a relaxation results from the scattering of an electron accompanied by the change in spin directions, which is possible due to the spin-orbit interaction. Since the electrons in the ferromagnetic are orbitally incoherent, the probability of scattering with spin change should not depend on the macroscopic spin polarization. The relaxation time  $\tau$  in the nanoparticle should coincide in order of magnitude with the characteristic time of remagnetization in a massive ferromagnetic domain. Naturally, the relationship  $N=N_++N_-$  remains unchanged.

In order to analyze the kinetics of remagnetization it is convenient to introduce the parameter *b*,  $(b=N_{-q}^{eq}/N_{+q}^{eq}<1)$  characterizing the equilibrium magnetization of nanoparticles. Then  $x^{eq} = (1-b)/(1+b)$ , and taking into account the relations (1) and (2) we obtain an equation for *x*:

$$\frac{dx}{dt} = \frac{I}{N} \left\{ \frac{\alpha - \alpha_p}{\alpha + \alpha_p} - \frac{1 - \alpha_p}{1 + \alpha_p} \right\} - \frac{1}{\tau} \left( x - \frac{1 - b}{1 + b} \right)$$
(4)

$$x(0) = x^{eq} = (1-b)/(1+b)$$
(4a)

A sufficient condition for remagnetization is determined by the value of the current. Specifically, remagnetization is achieved when the inequality x < 0 is satisfied under the influence of the current. Subsequently, the current can be switched off and the magnetization will be in equilibrium and opposite to the initial direction after the time duration $\tau$  due to relaxation. A sufficient condition for remagnetization follows from the Eq. (4) taking into account the Eq. (2), thus:

$$I > I_{cr} = \frac{N}{\tau} \frac{(1-b)(1+\alpha)}{(1+b)(1-\alpha)}$$
(5)

Let us estimate the value of the critical current  $I_{cr}$  in the expression (5). The iron oxide nanoparticle of size  $\sim 10^4$  nm<sup>3</sup> contains  $N \approx 10^5$  electrons, and the relaxation time  $\tau \approx 10^{-5}$  s [17,18]. Substituting these values into Eq. (5) we find that for  $\tau = 10^{-5}$  s remagnetization current  $I_{cr} \ge 5$  nA. This value is typical of the tunneling current in scanning tunneling microscope. The critical current is greater, the smaller the magnetization of the electrode and the greater the magnetization of the nanoparticles. At a current value that is slightly greater than  $I_{cr}$ , the remagnetization time is proportional to  $\tau$ , and it sharply decreases with increasing current [15].

Consider now the case when the magnetization directions of the ferromagnetic tips and nanoparticles are initially the same. Then it is necessary to change the direction of the current, i.e. the ferromagnetic tip should be the anode. The number of electrons with opposite spin directions coming from a non-ferromagnetic substrate in the nanoparticle will be the same. The flow of electrons with spin oriented along the tip magnetization from the ferromagnetic nanoparticles in the tip will be more than flow with the opposite spin direction (the effect of negative magnetoresistance [1,2]). As a result, the direction of the spin polarization of the nanoparticles under the influence of the current changes and becomes opposite to the direction of the ferromagnetic tip polarization. In this case, the kinetics of remagnetization is described by the equation,

$$\frac{dx}{dt} = \frac{I}{N} \left\{ \frac{1-\alpha_p}{1+\alpha_p} - \frac{1-\alpha\alpha_p}{1+\alpha\alpha_p} \right\} - \frac{1}{\tau} \left( x - \frac{1-b}{1+b} \right)$$
(6)

with the initial condition expressed in Eq. (4a). However, it is readily obvious that the criterion expressed in Eq. (5) for the magnitude of the critical current at which the nanoparticles remagnetize from the parallel magnetization of tip to the opposite magnetization remains the same.

# 3. Synthesis of magnetite nanoparticles

FeCl<sub>3</sub> and FeCl<sub>2</sub>salts (Fluka, Switzerland) were used without further purification. The Milli-Q water purification system (Millipore company, USA) was used to produce ultrapure water with a resistivity of 18 MOhm cm for all synthesis procedures. Magnetite (Fe<sub>3</sub>O<sub>4</sub>) nanoparticles were synthesized by the method of Massart [19]. The associated chemical reaction is expressed as follows:

$$2FeCl_3 + FeCl_2 + 8NH_4OH \rightarrow Fe_3O_4 + 8NH_4Cl + 4H_2O$$

$$\tag{7}$$

In order to provide the necessary ratio of Fe<sup>2+</sup> to Fe<sup>3+</sup>ions in the aqueous reaction system, aqueous solutions of FeCl<sub>2</sub> and FeCl<sub>3</sub> salts were mixed in the ratio of 2.5 ml 2 M FeCl<sub>2</sub> solution to 10 ml 1 M solution of FeCl<sub>3</sub>. Then 125 ml 0.7 M aqueous NH<sub>4</sub>OH solution was added to the resulting salt solution and the mixture was stirred vigorously for 30 min. Next, the magnetic fraction was extracted from the reaction mixture using a permanent magnet (samarium-cobalt magnet with field strength of about 1000 Oe) by the immobilization of magnetic nanoparticles at the wall of the vessel containing the reaction mixture. Thereafter, the resultant nanoparticles were washed with deionized water twice and placed in 125 ml of deionized water to form a suspension. A suspension of synthesized ligand-free colloidal magnetite nanoparticles was stabilized electrostatically by adding hydrochloric acid solution (the final pH value in the suspension of nanoparticles was 3.6–3.8). Finally, the resulting suspension of magnetite nanoparticles was subjected to ultrasonic treatment for 15 min.

The synthesized nanoparticles were characterized and studied by transmission electron microscopy (TEM) using LEO 912AB, IOME GA instrument and by dynamic light scattering technique using Brookhaven Instruments Corporation system 90 Plus. The results indicated an average size of the synthesized magnetite nanoparticles of about 10 nm (Fig. 1) and the nanoparticle electrostatic surface zeta potential ( $\zeta$ -potential) of +20.5 mV at pH=3.8.

The isoelectric point (the point of zero charge) of magnetite pI=6.8, so at aqueous suspension pH values lower than pH=6.8 the colloidal magnetite nanoparticles have a positive electrostatic charge, and can be stabilized effectively by the interparticle electrostatic repulsion. Deionized water with pH value in the range 3.6–3.8 was added to the suspension in order to change the concentration of the



Fig. 1. Characteristic electron microscopic image of synthesized colloidal magnetite  $Fe_3O_4$  nanoparticles. Image was obtained by transmission electron microscopy.

colloidal suspension of magnetite nanoparticles, in particular for dilution of the suspension with the aim of depositing individual magnetite nanoparticles onto the graphite surface.

In order to establish the structure of the synthesized nanoparticles the characteristic electron diffraction patterns at the nanoparticles were determined by TEM. Fig. 2 shows a characteristic image of the corresponding diffraction pattern. A comparison of the diffraction pattern from the present synthesized nanoparticles with that from a reference sample of magnetite nanoparticles demonstrates the coincidence of the positions of the diffraction reflections and their intensities. The result indicates that the synthesized nanoparticles are nanophase magnetite. Because the structural characteristics of the crystal lattice of the other magnetic iron oxide – magnetite  $Fe_2O_3$  – are very close to the lattice parameters of the magnetite, the presence of impurities of magnetic nanophase magnetite in the synthesized nanoparticles is not excluded.

## 4. Experimental conditions

Experiments were performed with STM to investigate the change of magnetization of iron oxide nanoparticles localized on

the surface of a conducting non-magnetic substrate. The STM tip was made of iron wire of diameter d=0.3 mm pre-magnetized in the field of a permanent magnet. The experiments were conducted in ultrahigh vacuum conditions with residual gas pressure in the STM chamber of  $P=2 \times 10^{-10}$  Torr. For this purpose aqueous suspension of iron oxide was deposited on the surface of highly orientated pyrolytic graphite (HOPG) and dried at room temperature. The sample prepared was placed in the STM chamber and heated up to T=600 K.

The standard methods of scanning tunneling microscopy and spectroscopy were used to study the morphological and electronic structures of the samples. The sample surface was scanned in constant current mode with tunneling current I=2-20 nA and voltage at the nanocontacts, (i.e. the potential difference between the STM tip and substrate), U in the range -2 V to +2 V. The magnetization of the nanoparticles and the change in magnetization were determined directly during scanning by the current–voltage dependence of the STM tunneling current, measured at different points on the sample surface both with bare tip surface and when covered with nanoparticles of iron oxide.

The following spectroscopic procedure was used to measure the current–voltage relationship. The STM tip was fixed above the sample surface and a voltage was applied to the STM nanocontact within the defined range, then the magnitude of the tunneling current was measured. The effect of GMR was used to determine the direction of the nanoparticle magnetization before and after passage through the nanoparticle, of strong polarized current greater than the critical value. The measurement was carried out using the following five steps.

- i. The current–voltage characteristic (CVC) of the current through the nanoparticle is measured for a sufficiently long distance between the STM tip and nanoparticle.
- ii. The tip is advanced towards the nanoparticle, and a strong polarized current is passed through the nanoparticle such that the polarized electrons jump from the tip to the nanoparticle. The tip is the cathode at this second step.
- iii. The tip is lifted up and the CVC is measured at weak currents. If the measured CVC turns out to be the same as that of step (i), then the magnetization directions of the tip and ferromagnetic nanoparticle were the same up to the second step.
- iv. The tip is again advanced towards the particle and a strong current is passed through the tip, but in the direction that is opposite to the stream of step (ii), i.e. the tip is now the anode and electrons jump from the nanoparticle to the tip.
- v. The tip is moved away from nanoparticle and the CVC is measured. If the resistance of the tip-nanoparticle tunnel junction significantly increases compared to the step (iii), then

Fig. 2. Electron diffractograms obtained from reference sample of standard colloidal magnetite nanoparticles (a), and nanoparticles synthesized in the present study (b). The numbers of crystallographic planes of the crystalline phase of magnetite causing the corresponding reflexes are indicated in the figure.

the magnetization directions of the tip and nanoparticle were opposite to one another after step (iv) according to GMR. In other words, a strong current turns over the direction of magnetization of ferrite nanoparticles. The above procedure is then repeated and a strong electron stream from the tip-cathode again turns the direction of magnetization of the nanoparticle.

Five film samples were used for each measurement in order to assess the repeatability of the data. The maximum relative error in the measurement of sensor response associated with hardware errors and irreproducibility of film composition during synthesis was 10%.

# 5. Results and discussions

Fig. 3a shows the topographic image of the HOPG surface of  $1.6 \times 1.6 \,\mu\text{m}$  size. The detailed profile of the sample within the indicator line at the lower part of Fig. 3a is presented in Fig. 3b. The sample under investigation consists of two closely spaced nanoparticles of iron oxide. The topographic measurements show that islands of irregular shape were formed on the surface of graphite with a maximum length of about 200 nm, consisting of nanoparticles with lateral size of less than 20–30 nm.

Using the procedure described in the previous section, let us start with the measurement of the current passing through the iron oxide nanoparticle. Fig. 4 presents the current-voltage relationship of the tunneling current of STM, measured during scanning by the magnetized iron tip with graphite surface area of  $50 \times 50$  nm size. Curve 1 corresponds to the area containing a single nanoparticle of iron oxide. For comparison, also presented on Fig. 4 is curve 2 showing the current–voltage relationship of the STM tunneling current measured at the HOPG area, which is free of iron oxide using the same scanning parameters (curve 2). These curves essentially differ both qualitatively and quantitatively. First, curve 2 corresponding to the HOPG area has an S-shape distribution which is typical for nanocontact with pure graphite. On the other hand, curve 1 corresponding to the nanoparticles of iron oxide is nearly linear. Secondly, the absolute value of the tunneling current for curve 2 is much smaller than that for curve 1. These data show that the conductivity of the tunneling junction formed by an iron tip and graphite is lower than the conductivity of the tunneling junction with iron oxide nanostructures.



**Fig. 4.** The current–voltage relationships measured on the iron oxide nanoparticles (curve 1) and graphite surface without nanoparticles (curve 2).

The following procedure was used to change the magnetic moment of the iron oxide nanoparticles. The value of the tunneling current was increased up to 30 nA while maintaining the voltages on the STM nanocontact (U = +2 V, i.e. ferromagnetic)STM tip was the anode). For this purpose the STM tip was advanced to the sample surface at a distance of  $\Delta Z \approx 0.1$  nm. Scanning of the sample surface was performed with an average speed  $V_{scan} \approx 140$  nm/s. Thus for the size of nanoparticles  $l \approx 20$  nm (Fig. 3), the duration of current flow through the nanoparticles was  $t = l/V_{scan} \approx 0.14$  s, which is comparable to the value of about 0.2 s, that was used in the SP-SPM method [13]. This time duration is the several orders of magnitude larger than the characteristic times of the nanoparticle remagnetization which was evaluated in a previous study [15]. Therefore the nanoparticles will certainly have time for remagnetization if the current is greater than the critical value. After the completion of the cycle, the tip was moved away from the sample surface to its initial position, and the surface scanning was repeated. The results of these measurements are presented in Fig. 5.

Fig. 5 shows that the shape and intensity of the current–voltage relationship corresponding to the island of iron oxide (curve 1)



Fig. 3. The HOPG surface containing nano-iron oxide nanoparticles: topographic image (a), and profile (b).



**Fig. 5.** The current–voltage relationships measured on the iron oxide nanoparticles (curve 1) and graphite surface without nanoparticles (curve 2) subsequent to electron tunneling current, I=20 nA,  $U_{bias} = +2$  V.

becomes closer to that of the HOPG area (curve 2), when graphite substrate is the source of electrons. A comparison of the system conductivity before exposure to strong current (curve 1 in Fig. 4) and after exposure (curve 1 in Fig. 5) shows that the conductivity decreases under the influence of strong current. In accordance with the theory of GMR [1,2] (see also works [20–22]), the conductivity decreases when the magnetization of the ferromagnetic electrodes becomes antiparallel. This implies that the magnetization of the STM tip and nanoparticle were approximately parallel before the influence of strong current, and become antiparallel after such exposure [15]. The approach of a ferromagnetic STM tip to the sample without the simultaneous increase of the tunneling current does not produce the effects described above. Thus the tip does not remagnetized the magnetic nanoparticle in this case.

It is possible for the magnetic moment vector of the nanoparticles to recover to the initial value, by passing strong current through the nanoparticle in the opposite direction [15]. For this purpose the procedure of STM tip advance to the sample surface was repeated, but the direction of the tunneling electron current was changed by changing the sign of the voltage on the STM tunneling nanocontact, i.e., the ferromagnetic tip becomes the cathode. The magnitude of the voltage was U = -2 V. The result is shown in Fig. 6.

Fig. 6 shows two current–voltage relationships of the STM tunneling current, measured on nanoparticles of iron oxide (curve 1) and the HOPG surface free from adsorbate (curve 2). Curve 2 does not change in shape and intensity while curve 1 (the current through the iron oxide) again has changed to be almost linear. In this case, as in Fig. 4, the absolute value of the tunneling current (curve 1) far exceeds the values of the tunneling current (curve 2) in the entire range of voltages considered. Thus, we can conclude that due to the procedure described above the direction of the magnetic moment of the iron oxide nanoparticles has returned to its initial state.

The threshold values of the tunneling current for the changing of magnetic moment direction of the iron oxide nanoparticle were also defined. For this purpose we used the scanning of the sample surface with a constant average speed ( $V_{scan} \approx 140 \text{ nm/s}$ ) and voltage U = +2 V applied to the STM nanocontact. In this case the value of the remagnetization tunneling current often referred to as "magnetic reversal" current [14] ranged from 2 to 20 nA with increments of 2 nA.

After every scan of the sample selected area the STM tip was



**Fig. 6.** The current–voltage relationships measured on the iron oxide nanoparticles (curve 1) and graphite surface without nanoparticles (curve 2) subsequent to electron tunneling current, I=20 nA,  $U_{bias} = -2$  V.

removed so that the tunneling current over clean graphite would return to the initial value (I=2 nA) at a voltage of 2 V and the current-voltage relationship of the tunneling current through the nanoparticle were measured in the range -2 V  $\leq U \leq +2$  V. The threshold current was defined as the value of the tunneling current corresponding to the change of shape and intensity from the type represented by curves 1 in Figs. 4 and 6, to the one represented by curve 1 in Fig. 5. Fig. 7 presents the dependence of the tunneling current on the absolute remagnetization current at U = +1 V measured after the change of the nanoparticle magnetic moment. The threshold tunneling current obtained is  $I_{thresh} \approx 9$  nA, which corresponds on the order of magnitude to the previously predicted values [15]. The heating of the sample can be neglected at such low currents especially because the experiment is performed at room temperature, and the sample temperature is considerably higher than that used in a previous study [14].

The measured threshold current is significantly smaller than that reported in the literature [12–14] by about 2 orders of magnitude, and the size of remagnetized ferromagnetic nanoparticle is larger than in the previous studies [12–14]. Such differences could be qualitatively explained as follows.



Fig. 7. Dependence of tunneling current on remagnetization current.

The number of polarized electrons in a nanocluster [12–14] is much smaller than in the  $Fe_3O_4$  nanoparticle considered in this study. Thus, the threshold current for repolarization nanocluster should be correspondingly smaller. However, it should be noted that the thickness of the iron nanoclusters is only a few angstroms [12–14], and the length of the spin-exchange electron scattering in polarized current on nanocluster electrons is much more. In other words, the flying spin-polarized electrons, for the most part, do not have time to interact with the electrons of the thin nanocluster and essentially skip through it. Therefore, a high specific current is needed for the spin turning since only a small part of the current spin polarization remains in a thin nanocluster.

The second equally important reason is that the characteristic spin relaxation time  $\tau$ in pure iron of the nanocluster [12–14] with very small coercive force is several orders of magnitude less than in the Fe<sub>3</sub>O<sub>4</sub> nanoparticles with a high coercive force, i.e., with a wide hysteresis loop. Thus, the characteristic time of domain remagnetization measured in a previous study [17,18], is on the same order of magnitude as  $\tau$  [15], but less than or about 10<sup>-8</sup> s in magnetization is proportional to  $1/\tau$  [15], and therefore it may be greater for the iron nanocluster than for nanoparticles of iron oxide.

# 6. Conclusions

An experimental study was used to prove the possibility to change the magnetic moment of large iron oxide nanoparticles by electric current between the ferromagnetic STM tip and a ferromagnetic nanoparticle on non-ferromagnetic (graphite) substrate. If the polarizations of STM tip and nanoparticles are oppositely directed, then in order to change the polarization of nanoparticle it is necessary that the larger number of electrons within the tip spin polarization enters the nanoparticle, and the larger number of electrons within the nanoparticle polarization leaves the nanoparticle. This implies that the tip should be a cathode and the nonferromagnetic substrate the anode. If the polarizations of nanoparticle and tip coincide, then in order to change the direction of nanoparticle polarization it is necessary that an increased flow of electrons with the spin directed to nanoparticle polarization leave the nanoparticle to the tip, and an approximately equal number of electrons with different polarization enters the nanoparticle from the substrate. This implies that the tip should be the anode and the substrate the cathode in this case.

The threshold value of the tunneling current was found to be equal to 9 nA. This threshold value for the nanoparticle size investigated ( $\sim 3.10^3$  nm<sup>3</sup> for nanoparticle height of about 15 nm and lateral diameter of 50 nm) corresponds to the values predicted in a previous study [15]. Such a value of critical current is substantially less than that established in other previous studies [12–14]. This discrepancy is attributed to two factors the case considered here. First, the larger time  $\tau$  of electron spin relaxation occurs because of spin–orbital interaction. Second, in the thick nanoparticle, full spin polarization of current flows over the spin system of nanoparticles.

It should be emphasized that the characteristics of interaction of polarized electrons with the nanoparticle spin system significantly depend on nanoparticle thickness. If the thickness of polarizable nanoparticle is small [12–14], focus is on the action of magnetic moment of polarized current on nanoparticle spin system. This moment acts like an external magnetic field, and the magnitude of field required to change the nanoparticle polarization depends on the magnetic anisotropy of the nanoparticle. In the case considered here involving a thick nanoparticle, the magnetic moment of polarized current, that is, non-collinear to magnetization of nanoparticle is of no consequence, since the thickness of nanoparticles is significantly larger than the path length of electron on which its spin state depends [15]. The overturning of the magnetization occurs through the non-equilibrium state occurring under the influence of the current, where the numbers of electrons with opposite spins are equal.

Based on the relation in Eq. (5), the critical current is proportional to  $\tau^{-1}$ . This value is determined by the spin–orbital interaction of electron with the crystal lattice of the nanoparticle, which causes the magnetic anisotropy of the ferromagnet. Thus, there is the obvious qualitative relationship: the larger the  $\tau^{-1}$ , the greater the crystal magnetic anisotropy of the ferromagnet and the larger the critical current magnetization reversal. The change of sign of the magnetic moment is determined by the change in conductivity at low currents, according to the theory of current in systems of ferromagnetic nanoparticles [20–22].

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