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Journal of Magnetism and Magnetic Materials 316 (2007) 236-239

www.elsevier.com/locate/jmmm

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Available online 3 March 2007

Abstract

We report unconventional oscillatory tunnel magnetoresistance as a function of applied bias in the magnetic tunnel junction with the Al₂O₃ barrier. We attribute this feature to the inhomogeneity of the potential barrier structure. Ferromagnetic grains inside the potential barrier are formed at the technological stage. In this case, the TMR oscillation occurs due to the discrete charging effect that is well known as the Coulomb blockade.

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PACS: 85.75.Bb

Keywords: Magnetoresistance osciilation; Magnetic tunnel junction; TMR

1. Introduction

The spin-dependent electron transport in magnetic tunnel structures consisting of two ferromagnetic metal layers separated by a thin nonmagnetic insulator (FM/I/FM structures) remains the subject of intensive research [1]. This is caused not only by the application potential of the tunneling magnetoresistance effect (TMR) in magnetic tunnel junctions (MTJs) for use in magnetic storage devices and magnetic sensors but also by the fact that new phenomena observed in the FM/I/FM structures represent the fundamental interest for researchers [2,3]. The primary aspects of the TMR effect are successfully described in the framework of the simple model that predicts the dependence of TMR on difference of density states (DOS) for two spin directions of an itinerant electron in the FM electrodes and on probability of transmission of tunneling electrons [4]. However, the analysis of the spin-dependent electron transport in real FM/ I/FM structures is complicated because it involves many parameters characterizing both an energy state of magnetic

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electrodes and a barrier structure, such as electronic structure of FMs, insulating layer and FM/I interfaces, disorder inside a barrier and FM electrodes etc. These parameters determine, in particular, the dependence of TMR on temperature (T)and bias voltage (V_{DC}). Experimental measurements of these dependences in various MTJs often give the controversial results. For example, some experiments demonstrate monotonous decreasing of TMR with increasing of T and $V_{\rm DC}$ [5,6], while others reveal an oscillatory behavior [7,8]. At present, many mechanisms responsible for TMR peculiarities in MTJs remain unclear, whereas an understanding of the origin of these mechanisms is of great importance for science and technology.

In the present paper, we report the experimental observation of an unconventional oscillatory behavior of the tunnel magnetoresistance MTJ as the bias voltage is increasing in the "exchange-biased" AFM/FM1/I/FM2 MTJ with the Al₂O₃ barrier (AFM indicates the antiferromagnetic layer).

2. Experimental details

MTJs with the complete layer sequence Ta(50 Å)/Cu(100 Å)/Ta(50 Å)/NiFe(20 Å)/Cu(50 Å)/Mn₇₅Ir₂₅(100 Å)/

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 $Co_{70}Fe_{30}(25 \text{ Å})/Al_2O_3(15 \text{ Å})/Co_{70}Fe_{30}(25 \text{ Å})$ were deposited onto thermally oxidized Si wafers by DC magnetron sputtering with ultra-clean Ar(9N) as a process gas at base pressure in a chamber of 3×10^{-9} Torr. Co₇₀Fe₃₀ alloy was used to form a "free" FM1 electrode (top electrode) and an outer FM2 electrode, the latter being "pinned" due to exchange bias field provided by an antiferromagnetic layer with composition Mn₇₅Ir₂₅. The Ta/NiFe/Cu stacks below Mn₇₅Ir₂₅ improved the MnIr plane FCC-(111) crystalline orientation [9], thus causing the exchange-coupling enhancement [10,11]. To form a barrier, metallic Al with the thickness of 15 Å was deposited and subsequently oxidized in the oxidation chamber with a radial slot antenna (RLSA) for 2.45 GHz microwaves generation [12]. Plasma oxidation was realized in the atmosphere of the inert gas Kr mixed with the molecular gas O2. The in-situ patterned junctions were prepared using a shadow mask during deposition; the iunction sizes were $180 \times 180 \,\mathrm{um}^2$.

The junction samples were thermally annealed at $250 \,^{\circ}$ C for 1 h in the magnetic field of 1 kOe, and then field cooled. The transport properties of the fabricated structure were measured by the conventional four-probe method.

3. Results and discussion

We have studied TMR of the fabricated junction as a function of the applied magnetic field (H) at various bias voltage $(V_{\rm b})$ in temperature range from 300 K down to 78 K. It appeared unexpected that with temperature decreasing the TMR effect in the junction either increases or decreases depending on a $V_{\rm b}$ value. Fig. 1 shows characteristic dependences of magnetoresistance on H (TMR(H)) measured at 78 K and 300 K for two fixed $V_{\rm b}$ values. TMR(H) was evaluated as $(R(H)-R_{\min}/R_{\min})$, where R_{\min} is the junction resistance at $H_1 = 2000$ Oe. At this field magnetization of a "free" FM layer of the structure is parallel to that of the outer (pinned) FM layer and, hence, the tunnel resistance has the minimal value. Drastic jump of the resistance below H = 0 is attributed to formation of the antiparallel alignment state, where magnetizations of two FM electrodes are antiparallel. Further decrease of the resistance with negative magnetic fields is determined by the remagnetization processes taking place in the FM electrode being exchange coupled with the AFM layer.

It is seen from the figure that the resistance peak broadens upon the junction cooling that indicates strengthening the exchange coupling between the AFM and "pinned" FM layers. At fixed temperature, the change of $V_{\rm b}$ gives rise to a change of the height of the resistance peak, at the same time, its shape remains invariable.

Since the pattern of the plot TMR(H) is nearly independent of V_b and there is only the TMR effect value change with V_b , we have measured the maximal value of the tunnel magnetoresistance (TMR ratio) as a function of V_b at various temperatures. TMR ratio was evaluated by difference between $I(V_b)$ curves at the applied fields H_1 and

Fig. 1. TMR ratio as a function of magnetic field at various temperatures. Junction voltage bias (a) $V_b = 1 \text{ mV}$; (b) $V_b = 7 \text{ mV}$.

 $H_2 = -150 \,\text{Oe}$ (see Fig. 1) that form parallel and antiparallel alignment junction states, respectively. Calculating the dependences of the junction resistance value from $I(V_{\rm b})(R = V/I)$, we determine the TMR ratio as TMR ratio = $(R(H_2) - R(H_1))/R(H_1)$. The $I(V_b)$ curves measured at temperatures 78 and 300 K and applied fields H_1 and H_2 are presented in Fig. 2(a). One can see the abrupt bends in the curves recorded at H_1 between 0–25 mV and 0–12 mV for 78 K and room temperature, respectively. The curvature of the $I(V_b)$ dependences is strongly modified at these features (or around them) by magnetic field change from H_1 to H_2 . Such modification of the $I(V_b)$ curves by applying magnetic field causes the abrupt changes of the TMR ratio in the range of $V_{\rm b}$ values where the features in the $I(V_{\rm b})$ dependences are observed. As a result, the oscillatory-type behavior of the TMR arises with $V_{\rm b}$ increasing. Fig. 2(b) shows the $V_{\rm b}$ dependence of the TMR ratio for two temperatures taken from the curves $I(V_{\rm b})$ recorded at H_1 and H_2 (Fig. 2(a)). It should be noted that the features in the $I(V_b)$ dependences and, consequently, in TMR ratio($V_{\rm b}$) dependences are observed starting from $V_{\rm b} = 0$ within the limited range of voltage





Fig. 2. (a) Voltage–current characteristic of the junction at temperatures 300 and 78 K in different magnetic fields. (b) TMR ratio as a function of the voltage bias for two temperatures 300 and 78 K.

bias on the junction. With further $V_{\rm b}$ increasing the TMR ratio smoothly decays. Besides, the $V_{\rm b}$ range where the oscillatory-type behavior of TMR are observed and also the character of the oscillations depend strongly on temperature. Indeed, as temperature decreases the $V_{\rm b}$ range broadens, while the amplitude of oscillations increases.

The observed non-linear voltage–current characteristics, their dependence on magnetic field value and, as a consequence, the TMR ratio oscillations cannot be explained in the framework of existing models of tunnel magnetoresistance under the condition of ideality of the studied MTJ. In general, an oscillation and even inversion of the TMR ratio may occur in a magnetic tunnel junction within an appropriate bias range, however, the calculations show that this behavior could be observed at the bias voltage much higher than that used in our experiments. Moreover, the period of the oscillations should be significantly greater than in our case, and the amplitude of the oscillations should strongly decrease with V_b due to suppressing of the spin-dependent tunneling effect at the high bias voltage.

In our case, the most probable origin of the nonlinear conductivity behavior is the formation of an inhomogeneous structure of the potential barrier (Al₂O₃ spacer) at the technological stage. We suggest the formation of ferromagnetic clusters with composition similar to that of magnetic electrodes (Fe-Co). In fact, the double ferromagnetic junction consisting of two magnetic electrodes with metallic ferromagnetic grains between them is formed that is separated from both electrodes by an insulating barrier. In this case, the TMR oscillations can be explained by a discrete charging effect known as the Coulomb blockade [13]. If the capacitance C of a grain is small enough for charging energy $e^2/2C$ to be the same order of magnitude as the thermal energy $K_{\rm b}T$, the effect caused by discrete charging is observed on the corresponding voltage-current characteristics. The current through the junction increases by a step at voltages where it is energetically favorable for an electron to tunnel to the grain. Occurrence of these current steps on the voltage-current characteristic is known as the "Coulomb staircase". The situation is complicated by the fact that the tunneling process will depend on mutual orientation of magnetizations of electrodes and magnetic grains inside the barrier of the structure. Besides, it should be expected that a large number of grains are involved into the tunneling process, with rather broad distribution in sizes and distances between grains and electrodes. All mentioned results in the relatively complex Coulomb blockade effect that cannot be easily analyzed. To verify our suggestion, the additional experiments have to be carried out on structures with purposely created inhomogeneities with different composition and sizes inside a potential barrier.

4. Conclusion

We attribute the nonlinear conductivity and oscillations of the TMR ratio at increasing of bias of the magnetic tunnel junction under study to the inhomogeneity of the potential barrier structure, or, in other words, to the features of the technology used for junction fabrication. Having analyzed the experimental data, we suggest that the potential barrier of the structure consists of the insulator layer and small ferromagnetic metal particles imbedded in it, whereas the observed magnetoresistance oscillations are related to the Coulomb blockade phenomenon.

In our opinion, the investigations similar to those presented in this paper are of great importance as they facilitate advancement in understanding the mechanisms of the electron spin-dependent transport in various solid-state systems and thus open new possibilities for creation of the next-generation of magnetic structures with the controlled spin-dependent current.

Acknowledgments

This work was supported by the Korean Science and Engineering Foundation (KOSEF), Grant no. R01-2004000-10882-0. N.V. Volkov thanks the Russian Science Support Foundation for support.

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