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Percolation-related magnetic coupling and magnetoresistance properties in $Fe/Si_{1-x}Ag_x$ multilayers*

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In this paper, we present a study of the magnetic coupling and magnetoresistance (MR) properties in $Fe/Si_{1-x}Ag_x$ multilayers with a granular $Si_{1-x}Ag_x$ spacer layer. We have found that, with increasing silver content (x) in a silicon matrix, the magnetic state of multilayers changes from a nonmagnetic coupling state to weak antiferromagnetic around the percolation point of the ~2.4 nm thick spacer $Si_{1-x}Ag_x$. The MR measurements also reveal an abrupt increase of MR near the same percolation point. These changes are ascribed to the formation of the percolation path in the granular spacer.

Keywords: interlayer coupling, doping, magnetoresistance
PACC: 7570L, 6170T, 7215G

The discovery of the oscillation of magnetic coupling through a transition metallic spacer layer[1] and the corresponding giant magnetoresistance (GMR)[2] in antiferromagnetic (AFM) configuration between magnetic layers has opened a new exciting research area in modulated nanostructures, promoting a revolutionary development in information storage and magnetic sensors. The common feature of these modulated nanostructures is that the interlayer coupling and the magnetoresistance (MR) values oscillate with the thickness of the spacer layer.[3–5] Moreover, the coupling strength and periods can also be mediated by the topology of the Fermi surface using a metallic alloy as the spacer layer or alloy magnetic layer with different composition ratios, such as Fe/CrV/Fe, Co/CuNi/Co and Fe-Co-Ni/Cu.[6]

Recently, an AFM configuration in Fe/Si multilayers (MLs) reported by Toscano *et al.*[7] has aroused great interest from the scientific community.[8–13] Experiments have revealed the heat-induced or photo-induced changes of interlayer coupling from ferromagnetic (FM) to AFM, which could be explained by excitation of carriers into the conduction band of Si that makes the communication of spin information between the iron layers possible.[9] Negative MR was also observed and two types of temperature dependence were found as a function of Si thickness.[11]

In this paper, we present a novel variant of magnetic coupling and MR in Fe/Si MLs with silver doping in the Si spacer layer. It is known that in Fe/Si MLs, when the thickness of the spacer layer is greater than ~2 nm, an α-Si spacer layer is formed which leads to a nonmagnetic (NM) coupling (or weak ferromagnetic exchange coupling as indicated by Briner *et al.*[9]) state between the iron layers for lack of spin carriers in the spacer layer.[7,8,11–13] Silver was chosen to be the doping element because of the absence of alloy phases between silver and silicon even at several hundred degrees.[14,15] Thus, a granular $Si_{1-x}Ag_x$ spacer is formed between Fe layers in our system. We have found that, with an increasing silver content (x) in the silicon matrix, the magnetic state of the MLs varies from NM to weak AFM coupling around the percolation point of the ~2.4 nm thick granular spacer $Si_{1-x}Ag_x$. The MR measurements show an abrupt increase of MR near the same percolation point. We

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ascribe these variations to the formation of the percolation path in the granular spacer. To our knowledge, this is the first report on the interlayer coupling and GMR associated with the percolation process in the granular spacer layer.

The $Fe/Si_{1-x}Ag_x$ MLs were grown at ambient temperature on water-cooled glass substrates by magnetron sputtering. The background vacuum was better than 2×10^{-5}Pa and the pressure of sputtering Ar gas was 1.0Pa. The composition ratio of silicon and silver in the spacer layers was controlled by adjusting the area ratio of the elements, taking into account the sputtering yields of the elements. The composition of the standard 300 nm thick $Si_{1-x}Ag_x$ layer was determined by electron-probe microanalysis (EPMA). The deposition rates of individual layers of MLs were always kept at $0.1\,nm\cdot s^{-1}$. The thickness of each layer was controlled by sputtering time through a preset computer program. Small angle x-ray diffraction (XRD) was used to investigate the multilayer structures, which showed good quality of prepared MLs. The magnetic properties were characterized using an alternating gradient force magnetometer (AGM) with a maximum in-plane magnetic field of 1.5 T. The standard four-probe method was used to measure MR at room temperature with the field in the film plane.

Here we give a brief description of Fe/Si MLs. The ratios of the remanence M_r to the saturated magnetization M_s and the saturation field H_s at room temperature for the $[Fe(3.0\,nm)/Si(t\ nm)]_{20}$ MLs are measured. There is a peak in H_s and a minimum in the remanence ratio for $t_{Si}\approx1.1$ nm, suggesting an AFM coupling between Fe layers. Both H_s and the remanence ratio are nearly constant after $t_{Si}>1.8$ nm. These results are in agreement with those of Endo *et al.*[13] From the characteristics of the saturation field H_s and remanence versus t_{Si}, the interlayer coupling between adjacent Fe layers could be summarized as: FM coupling for $t_{Si}<0.8$ nm, AFM coupling for $0.8<t_{Si}<1.8$ nm, and NM coupling for $t_{Si}>1.8$ nm for lack of the spin communication carriers in α-Si spacer layer, as indicated by Endo *et al.*[13]

The investigation on the silver doping effect on the magnetic coupling and MR was carried out in a wide thickness range of the spacer layer with a fixed 3 nm thickness of the iron layers. Here, we focus on three typical series of samples with different spacer layer thicknesses: 1.9 nm (α), 2.4 nm (β) and 3.5 nm (γ). Clearly, there is no coupling between the Fe layers with such spacer layer thicknesses for pure $[Fe(3.0\,nm)/Si\ (t_{si}\ nm)]_{20}$ MLs, as we have found and discussed above.

However, we observed a significant variation in the shape of magnetic hysteresis loops in the β series of samples, i.e. $[Fe(3.0\,nm)/Si_{1-x}Ag_x\ (2.4\,nm)]_{20}$ MLs, with increasing silver content (x). H_s and remanence ratios obtained from the magnetic hysteresis loops are plotted in Fig.1. Note that, only for the β series of samples, H_s and remanence ratio values (presented in Fig.1(a)) show a sudden change at about x=0.35, where H_s increases from ~0.020 to ~0.095T and the remanence ratio drops from ~0.75 to ~0.30. This obviously corresponds to a coupling transition between the Fe layers from NM coupling to AFM coupling at about x=0.35 with increasing silver content. In contrast, there are no obvious changes in magnetic hysteresis loops in the α and γ series, as shown in Fig.1(b), where H_s and the remanence ratio change only slightly, showing that there is no AFM coupling between the Fe layers in these samples.

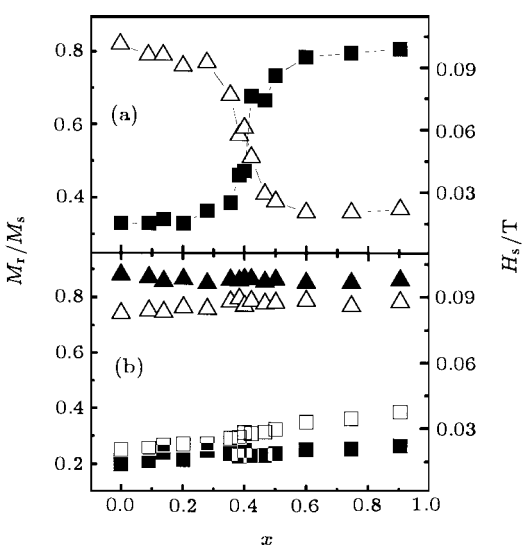


Fig.1. (a) Remanence ratio (M_r/M_s) (△) and saturation field (H_s) (■) as a function of silver volume x in $[Fe(3.0\,nm)/Si_{1-x}Ag_x\ (2.4\,nm)]_{20}$ MLs. (b) M_r/M_s and H_s as a function of silver volume x in $[Fe(3.0\,nm)/Si_{1-x}Ag_x\ (1.9\,nm)]_{20}$ (△ for M_r/M_s and □ for H_s) and $[Fe(3.0\,nm)/Si_{1-x}Ag_x\ (3.5\,nm)]_{20}$ (▲ for M_r/M_s and ■ for H_s) MLs.

The appearance of the AFM interlayer coupling in the β series of samples is quite different from that of samples with a metallic alloy as the spacer layer, in which the change of external spanning vector of the Fermi surface in the spacer layer corresponds to the variation of the interlayer coupling. It is well known that silver does not alloy with silicon and will form

a granular film in the co-sputtering process.[14,15] For the β series of samples, the silver particles could only offer large localized states in the forbidden energy gap of an α-Si matrix when the silver content is smaller. With an increasing silver content, the microstructure of $Si_{1-x}Ag_x$ changes from nanosized silver particles embedded in the α-Si matrix to larger silver crystallites in contact with each other. With the latter microstructure, the percolation path can be formed and the magnetic layers can thus communicate effectively with each other through this spin-carrier path by electrons. It is also interesting to note that the second AFM coupling peak in many metallic MLs, such as Fe/Ag/Fe films, only occurs around the spacer thickness of 2.3–2.5 nm,[16] which is almost the same as the spacer thickness range of our β series of samples. Actually, no magnetic coupling was found in the α or γ series whose spacer thickness is either significantly smaller or larger than ~2.4 nm, which is not in the second AFM peak coupling range for Fe/Ag/Fe films. Thus we suggest that the formation of the percolation path could be the reason for the change of magnetic coupling in the β series of samples.

We also note that the change of the microstructure of $Si_{1-x}Ag_x$ due to silver doping might also have an influence on the interfaces, and therefore the magnetic coupling. Since we have not found any magnetic coupling variation in the α and γ series of samples, which may have the same microstructure change due to doping, we believe that the AFM coupling found in the β series of samples is mainly due to the formation of the spin communication path.

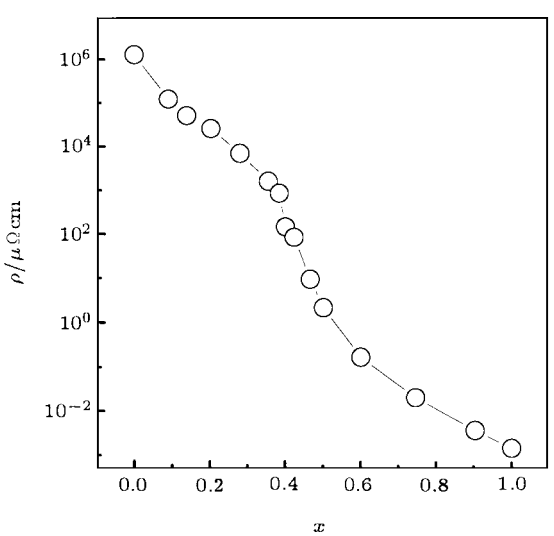


Fig.2. Zero-field resistivity of 300 nm $Si_{1-x}Ag_x$ thin films versus volume fraction x of silver at room temperature.

For a better understanding of the observed percolation-related magnetic properties in the β series of samples, we have measured the transport properties of the standard 300 nm $Si_{1-x}Ag_x$ films for reference. Since silver does not alloy with silicon, granular films always formed. Knowledge of such a metal-insulator granular system has been accumulating for quite a long time, especially about the percolation effect, which is an important physical phenomenon in this system.[17,18] Recent work on $Si_{1-x}Ag_x$ granular films have shown percolation-related optical properties.[14,15] Here, in our study, the resistivity of $Si_{1-x}Ag_x$ thin films under zero magnetic fields as a function of x at room temperature is presented in Fig.2. It can be seen that while the resistivity increases logarithmically with decreasing x in a wide range of x, a discontinuous increase can also be seen when x is in the range of 0.35–0.50. This indicates that the percolation threshold is around 0.4, at which the mechanism of the electrical conductivity is changed. A percolation threshold value of 0.3~0.5 for granular composite media is generally predicted by percolation theories.[17,18]

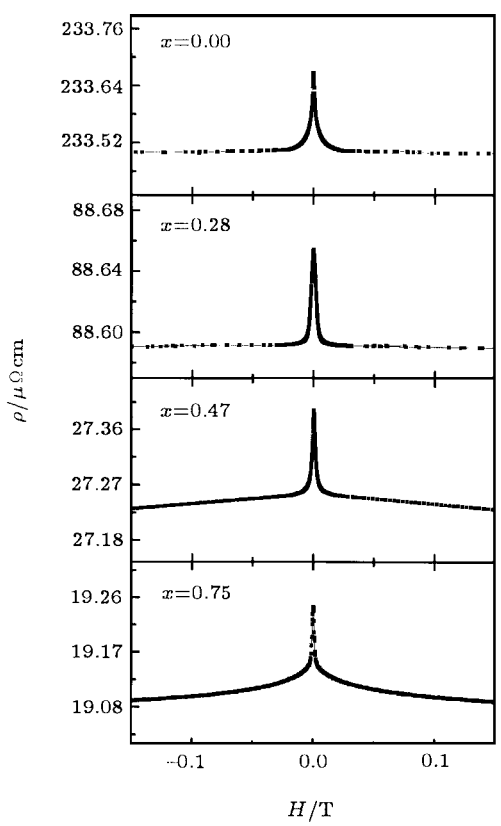


Fig.3. Four typical MR plots of [Fe(3.0 nm)/$Si_{1-x}Ag_x$ (2.4 nm)]$_{20}$ MLs with different volume contents of silver: x=0.00, 0.28, 0.47 and 0.75.

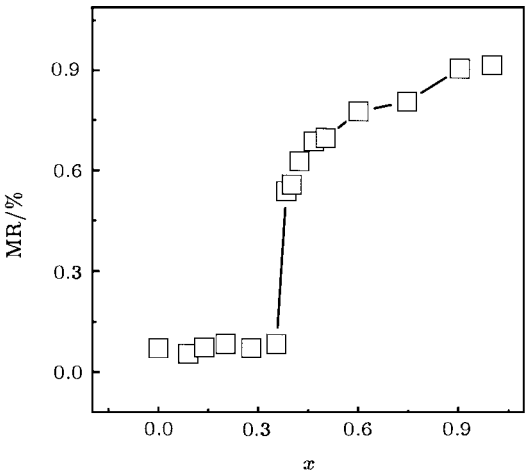


Fig.4. MR values of $[Fe(3\,nm)/Si_{1-x}Ag_x\ (2.4\,nm)]_{20}$ MLs versus volume fraction x of silver at room temperature.

To give further evidence in support of our above suggestion that the change of the magnetic coupling in the β series of samples may be due to the formation of the percolation path, we have also measured the MR properties of the β series of samples.

The typical MR curves in the β series measured at room temperature are plotted in Fig.3. Fig.3(a) plots the MR of the $[Fe(3.0\,nm)/Si(2.4\,nm)]_{20}$ ML sample. The MR value versus t_{Si} in pure $[Fe(3.0\,nm)/Si(t_{si}\ nm)]_{20}$ MLs are also measured. These results are in agreement with those of Inomata *et al.*[11] except for the behaviour of the MR value for $t_{Si} > 2$ nm. In our experiment, the MR value remained almost constant when $t_{Si} > 2$ nm whereas a minimum MR was obtained around $t_{Si} \approx 2$ nm in Ref.[11]. The difference may have arisen from the different preparation methods. Three typical MR plots of $[Fe(3.0\,nm)/Si_{1-x}Ag_x(2.4\,nm)]_{20}$ MLs with different volume contents of silver x=0.28, 0.47 and 0.75 are presented in Figs.3(b), 3(c) and 3(d), respectively. Compared to the MR value of pure Fe/Si MLs in Fig.4(a), the MR value of the sample with x=0.75, as shown in Fig.3(c), is increased by almost one order of magnitude. The MR value versus x for the β series of samples is shown in Fig.4. The most interesting observation is that a large abrupt increase of the MR value occurs around x=0.38, which is the same as the magnetic coupling transition point discussed above. We believe that the large jump of the MR value is due to the AFM coupling between the magnetic layers. The trend of MR versus x gives further support for the formation of the percolation path in the spacer layer. Here, we also note that the change of the MR value is sharper than that of the parameter of magnetic coupling near the percolation point. A better understanding of the microstructure and the density of localized states around the Fermi energy might be the key to explaining the difference.

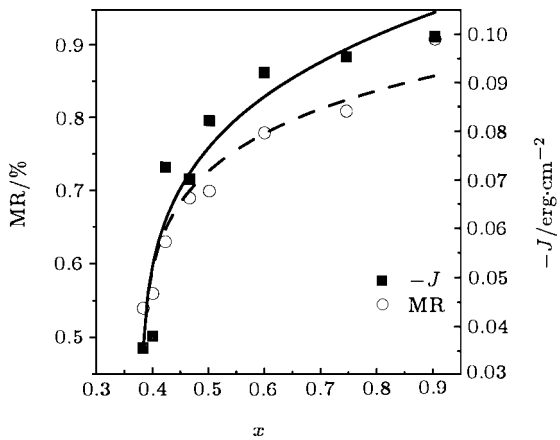


Fig.5. Dependence of MR and interlayer coupling strength J on the silver volume fraction x for $[Fe(3\,nm)/Si_{1-x}Ag_x\ (2.4\,nm)]_{20}$ MLs. The solid and dashed lines show the simulated results using Eq.(1) for J and MR, respectively.

In a metal-insulator granular system, the percolation threshold of the metal is also a conductivity threshold. Associated with the threshold is the well-known critical behaviour of the conductivity σ in the vicinity of the percolation threshold,[19,20]

$$\sigma = \sigma_0(p - p_c)^t, \tag{1}$$

where p is the content of the metal, and p_c is the threshold value. The value of t is 1.0~1.2 for 2D and 1.4~1.7 for 3D. σ_0 is a constant.

Here the AFM coupling strength J and MR for the β series of samples, when x is larger than the percolation threshold value 0.38, are also simulated with Eq.(1) and presented in Fig.5. We have obtained t=0.2 for J and t=0.1 for MR through the simulation. These values are much smaller than that for normal conductivity, which may be due to the different mechanisms of normal conductivity and MR. We note that MR and J in our samples increase more sharply when x is just above the threshold value, and increase slowly when x is increases. This may correspond to the fact that the GMR effect associated with the AFM interlayer exchange coupling in MLs formed by FM layers and NM spacers is due to the spin asymmetry of the reflection of the spin-dependent electrons at the interfaces. This means that as soon as the percolation path is formed in the spacer, although only few electrons could pass through the spacer at this stage, the

scattering asymmetry of the spin-dependent electrons could also cause a large MR effect, which can be seen in Fig.5.

In conclusion, we have presented for the first time experimental evidence for the percolation-related AFM interlayer coupling in $Fe/Si_{1-x}Ag_x$ MLs at room temperature. The concurrence of the abrupt jump of magnetic coupling and MR value at the percolation point of the $Si_{1-x}Ag_x$ spacer indicates that the formation of the spin communication path in the spacer strongly influences the interlayer coupling and MR in $Fe/Si_{1-x}Ag_x$ MLs.

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