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## MAGNETISM AND FERROELECTRICITY

# Magnetic Properties of Fe/Si/Fe Trilayer Films

G. S. Patrin\*, \*\*, S. G. Ovchinnikov\*, \*\*, D. A. Velikanov\*\*, and V. P. Kononov\*\*

\*Krasnoyarsk State University, Krasnoyarsk, 660041 Russia \*\*Kirensky Institute of Physics, Siberian Division, Russian Academy of Sciences, Akademgorodok, Krasnoyarsk, 660036 Russia e-mail: pat@iph.krasnoyarsk.su Received January 10, 2001

**Abstract**—The magnetization of Fe/Si/Fe trilayer films is experimentally investigated at low temperatures. It is found that the shape of the magnetization curves measured at T < 30 K depends on the thermomagnetic state of the system. The possible mechanisms of the interaction between iron layers are discussed. © 2001 MAIK "Nauka/Interperiodica".

## 1. INTRODUCTION

In recent years, multilayer magnetic systems have attracted the particular attention of many researchers, because they can be successfully used in the preparation of magnetic materials with controlled properties [1]. This is achieved by the proper choice of the materials of the main magnetic and intermediate layers, their thicknesses, and packing modes. The use of a semiconductor material as an intermediate layer provides a means for controlling the properties of these layers through external factors (impurities, different types of radiation, temperatures, fields, etc.).

A distinctive feature of magnetic films with a silicon interlayer (of thickness  $t_{Si}$ ) is that they can be rather easily obtained using different methods and, what is more important, possess unusual properties in readily attainable ranges of controlling parameters. In particular, Toscano *et al.* [2] found that, at T > 40 K, the exchange interaction parameter varies with temperature. Mattson et al. [3, 4] revealed a photoinduced change in the interlayer exchange parameter with the use of the Kerr effect. In our recent work [5], we studied the dependence of the internal effective magnetic field on the thickness of a silicon interlayer by electron magnetic resonance. It was demonstrated that, at  $T \ge 80$  K, the photoinduced contribution to the magnetic interaction between the iron layers in films at  $t_{Si} > 10$  Å has an antiferromagnetic nature.

In the present work, we investigated the low-temperature behavior of the magnetization of Fe/Si/Fe films in weak magnetic fields by the superconducting quantum interference device (SQUID) method.

## 2. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUE

The Fe/Si/Fe trilayer films were prepared by molecular beam epitaxy with an Angara setup, which was specially adapted for evaporation of magnetic materials [6]. Four samples with different thicknesses of the silicon interlayers were prepared in one evaporation cycle. For all the studied samples, the iron layer thickness  $t_{\text{Fe}}$  was equal to 50 Å. The thickness was checked against a quartz thickness meter and the growth rate of the film. One reference film with thickness  $t_{\text{Fe}} = 100$  Å was prepared in each evaporation cycle. The intensities of microwave absorption in the films were compared with the aim of revealing the difference between the masses of the magnetoactive materials. The magnetization was measured on a SQUID magnetometer [7]. The magnetic field was applied along the film plane.

### 3. RESULTS AND DISCUSSION

All the previous experiments [2–4] were performed in magnetic fields of the order of 1 kOe. In these works, it was assumed that the main mechanism responsible for the observed effects is associated with a change in the charge carrier concentration in a semiconductor interlayer either with a variation in temperature or under illumination. At the liquid-nitrogen temperature under equilibrium conditions, all conduction electrons in silicon are frozen. In this case, the signals taken from the iron layers indicated a nearly independent ferromagnetic behavior of these layers. According to our earlier measurements of the magnetic resonance parameters [5], the observed shift of the resonance field is caused by oscillations of the parameter of the exchange interaction between the iron layers and its value depends on the thickness of the silicon interlayer in the studied film (Fig. 1). The iron layers interact ferromagnetically at  $t_{Si} < 10$  Å and antiferromagnetically at  $t_{\rm Si} > 10$  Å. This behavior is observed at temperatures of 300 and 80 K. Note that, although the effect in the latter case is considerably weaker, all the main features are well reproduced.

Figure 2 shows the temperature dependences of the difference between the magnetizations  $\delta\sigma(T) = \sigma(T, 0) - \sigma(T, 0)$  $\sigma(T, t_{Si})$  for the reference and studied Fe/Si/Fe films at different silicon interlayer thicknesses. These dependences were measured in a magnetic field of 250 Oe upon heating of samples preliminarily cooled under different conditions. The applied field (250 Oe) is high enough for the samples to be at saturation at all temperatures. First, it should be noted that, in the temperature range 30–300 K, the dependences of  $\delta\sigma$  on  $t_{\rm Si}$  at T =const correlate with the curves depicted in Fig. 1. These results do not depend on whether the samples were cooled with or without a magnetic field. Second, the curves obtained under different cooling conditions considerably differ at temperatures below 30 K. It is also worth noting that the dependence of  $\delta\sigma(H_c = 250 \text{ Oe})$ - $\delta\sigma(H_c = 0 \text{ Oe})$  (where  $H_c$  is the magnetic field applied to the sample during its cooling) on the silicon interlayer thickness  $t_{Si}$  at a constant temperature correlates with the dependences displayed in Fig. 1.

For the film at  $t_{Si} = 20$  Å, which is characterized by the maximum antiferromagnetic coupling between the iron layers, the magnetization curves obtained at 4.2 K upon an increase (curve 1) and decrease (curve 2) in the magnetic field are plotted in Fig. 3. In this experiment, we measured the signal from the film on a substrate, then removed the magnetic film, and measured the signal from the substrate. The difference between these signals was taken as the magnetization of the studied film. It is clearly seen from Fig. 3 that an increase in the magnetic field leads to the appearance of two characteristic points in the magnetization curve:  $H_1 \approx 160$  Oe (the inflection point in the initial portion of the magne-tization curve) and  $H_2 \approx 240$  Oe (the point where the magnetization reaches saturation). In the case when the magnetic field decreases, no features are revealed at the field  $H_1$ . Since the magnetization in the given experimental geometry should be aligned with the film plane, curve 1 can be realized in the presence of crystalline magnetic anisotropy competing with the shape anisotropy. In the framework of the two-sublattice antiferromagnet model, this behavior becomes possible either in the case when the anisotropy axis is perpendicular to the film plane or in the presence of in-plane anisotropy. Therefore, the magnetic field  $H_1$  can be considered a field of the spin-flop state and the field  $H_2$  can be treated as a field attained after collapse of the sublattice. The magnetization behavior observed with a decrease in the magnetic field can be interpreted as a manifestation of either the ferromagnetism induced by the magnetic field or the magnetic-field-induced metastable state in which the magnetic anisotropy can be observed only after overcoming a certain energy barrier.

All these findings seem unexpected under the assumption that the interaction between the iron layers can be provided only by conduction electrons in the bulk of the silicon interlayer, because the carriers



**Fig. 1.** Dependences of the normalized shift of the molecular field on the silicon interlayer thickness for Fe/Si/Fe films at T = (1) 80 and (2) 300 K.



**Fig. 2.** Temperature dependences of the change in the magnetization per unit area  $\delta\sigma(T, t_{Si}) = \sigma(T, 0) - \sigma(T, t_{Si})$  at different silicon interlayer thicknesses  $t_{Si}$ : (1) 5, (2) 10, (3) 20, and (4) 30 Å. The open and closed symbols correspond to cooling in magnetic fields H = 0 and 250 Oe, respectively.

responsible for this interaction should not occur at low temperatures.

In order to explain the low-temperature behavior of the magnetization of the Fe/Si/Fe films under investigation, it is necessary to reveal the carriers responsible for the interaction between the iron layers and to elucidate the mechanism of this interaction. It is known that a transition region of the metal silicide–silicon type is formed at the metal–semiconductor interface [8]. According to the data of Auger electron spectroscopy,



Fig. 3. Dependences of the reduced magnetization on the magnetic field upon (1) increase and (2) decrease in the field. T = 4.2 K.

silicon comes into contact with d elements when the thickness of this transition region lies in the range from several to twenty angstroms (for example, 7-14 Å for Ni and approximately 3 Å for Pd). Imada et al. [9] demonstrated that the FeSi compound is a nearly ferromagnetic semiconductor. This means that FeSi possesses both weak metallic properties according to the magnitude of the electrical conductivity and semiconductor properties as judged from the temperature dependence of the electrical conductivity. The point is that the density of states of FeSi is not high enough for this compound to possess highly ferromagnetic properties. However, owing to the electron transfer from the metal, a region enriched with electrons is formed at the metalsemiconductor interface on the semiconductor side. It is easy to estimate [8] that, in the case when the total thickness of the silicon interlayer is of the order of 10 Å, the overlapping transition regions formed at the boundaries with both iron layers should be located throughout the bulk of the interlayer. In this case, the electron density turns out to be high enough for the ferromagnetic order to arise in the system. When the order in the system is formed according to the spin-density wave scheme, a further increase in the interlayer thickness leads to a change in the magnetic order. Apparently, the interlayer thickness  $t_{Si} \sim 20$  Å can be treated as an oscillation period in the system with a low density of states. A further increase in  $t_{Si}$  results in disturbance of the long-range order in the system due to the low density of states.

At low temperatures, the possibility of the interaction occurring through the states of band tails in the semiconductor should not be ruled out. As was noted by Toscano *et al.* [2], the silicon interlayer prepared through evaporation is in an amorphous state and the short-range order region is approximately equal to 16 Å. In this situation, the silicon interlayer contains a large number of defects and the electron transfer processes should be considered with due regard for the band-tail states [10]. In amorphous silicon, the mobility of charge carriers increases at temperatures below 30 K. The activation energies of these processes fall in the range 1-2 meV. Consequently, the interaction between the iron layers can proceed through the bandtail states. A decrease in the concentration of carriers with a decrease in the temperature can be compensated for by an increase in their mean free path. This mechanism can provide a memory effect in the spin state of the layer already vacated by electrons. In order to determine the specific mechanism of formation of a longrange order in the trilayer system, it is necessary to investigate in more detail the electronic properties at the iron-silicon interface and to elucidate the role of the band-tail states in charge carrier transfer.

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