

Preparation of heterogeneous film structures by thermally activated mass-transfer

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Electrical and magnetic characteristics have been studied in heterogeneous film structures Cr(25 nm)/(Al + Mn)(50 nm)/Cr(25 nm) and Fe(100 nm)/(Al + Mn)(200 nm)/Fe(100 nm) obtained using thermally activated mass transfer. The $dR(T)/dT$ dependence for the annealed film has shown a complex phase composition. The characteristic peak at T = 165 K corresponds to T_C (Curie point) of the Fe_2AlMn alloy. The results obtained show the principal preparation possibility of Heusler type heterogeneous film structures using thermally activated mass transfer.

Исследованы электрические и магнитные характеристики пленочных гетерогенных структур Cr(25 нм)/(Al+Mn) (50 нм)/Cr (25 нм) и Fe (100 нм)/(Al + Mn) (200 нм)/Fe (100 нм), полученных с помощью термически активированного массопереноса. Зависимость $dR(T)/dT$ показала сложный фазовый состав пленки после отжига. Характерный пик при температуре 165 K соответствует T_C (точке Кюри) сплава Fe_2AlMn. Полученные результаты свидетельствуют о принципиальной возможности создания пленочных гетерогенных структур гейслеровского типа с помощью термически активированного массопереноса.

The progress in the new field of electronics, spintronics, demands development of effective and reliable preparation methods of heterogeneous film structures. Such structures are used, e.g., in data readout from hard disks. The tunnel junction sensor includes a ferromagnetic film with a high degree of spin polarization, nonmagnetic, and antiferromagnetic layers. As antiferromagnetic layer, films of Cr–Al–Mn system were successfully used in spintronic devices [1]. The properties of the films made of such materials depend heavily on the accuracy of the stoichiometric composition. It is not always possible to achieve that by using known methods of material condensation on substrates. A more precise control of the final heterogeneous film composition is possible when using the interpenetration of separately deposited components of the preset composition. Such an interpenetration can be provided using thermally activated diffusion or mass transfer under laser pulse action. This work presents the electric and magnetic resistance study results of film structures obtained using thermally activated mass transfer.

To obtain the specified composition of the final film, the Cr (25 nm), Al + Mn (50 nm) and Cr (25 nm) layers were sequentially deposited on the glass ceramic substrates using electron beam in the vacuum chamber of double electron gun system VUP-5m. The interpenetration of the film components was provided by diffusive thermal annealing in vacuum ($5 \cdot 10^{-5}$ Torr) during the estimated time directly in the chamber of deposition system. The mass transfer

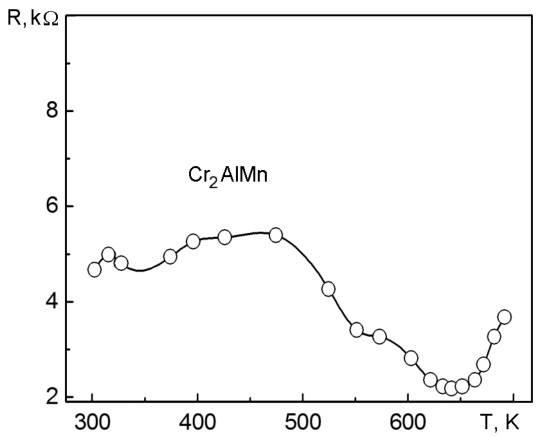

Fig. 1. Temperature dependence of the electrical resistance during the heating of a Cr–Al–Mn film structure of 100 nm initial thickness and formation of the Cr_2AlMn-film.

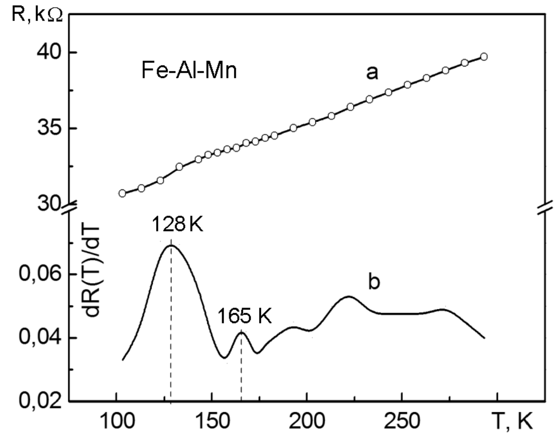

Fig. 2. Fe–Al–Mn film (400 nm initial thickness) electrical resistance vs temperature: $R = f(T)$ (a); $dR(T)/dT = f(T)$ (b).

in the film during the annealing was monitored by measuring the electric resistance (see Fig. 1). The annealing duration t was calculated using the known interdiffusion coefficients D of the components over the preset depth h as

$$t \approx h^2/D, \text{ where } D = D_0\exp(-E_a/kT).$$

As it follows from the dependence presented in Fig. 1, the resistance decreases monotonously in the temperature range from 473 to 643 K. At a further temperature increase, the main contribution to the resistance change is due to diffusion processes. According to [2], increasing content of Cr, as well as Mn, in Al causes an increase of the Al resistivity, as is observed in Fig. 1. That is why the annealing temperature of 623–673 K was chosen to provide the effective interpenetration of the film components. At higher temperatures, the effective formation of intermetallides is also possible [2]. The diffusive annealing duration t (1 hour) was chosen according to the interdiffusion coefficients of Mn–Al ($D \sim 10^{-11}$ cm^2/s at 623 K) and diffusion parameters of Cr in Al ($D_0 = 3.01\cdot10^{-7}$ cm^2/s, $E_a = 64.4$ kJ/mole) [7]. This provides the interpenetration of the film components to the depths of $h > 50$ nm in selected temperature range.

To check the application possibility of the diffusion method in preparation of the Heusler type heterogeneous films, the multilayered film of Fe(100 nm) – (Al + Mn)(200 nm) – Fe(100 nm) was annealed. After a rather short (30 min) diffusive annealing (T = 573 K), the film resistance was decreased from 150 to 49 Ohm at a slight increase of magnetoresistance (MR = 0.05 %) in the field H = 4 kOe. Such low magnetoresistance is explained by fact that annealed bulk Fe_2AlMn shows a magnetic transition below approximately 165 K [3].

Thus, to examine the formation possibility of the Fe_2AlMn phase, it was necessary to study the temperature dependence of the obtained film resistance in the 300–100 K range. To exclude possible errors connected with the inhomogeneous film cooling, all measurements were carried out at the absence of the temperature gradient along the film surface. To that end, the sample with the attached thermocouple (copper-constantan) was gradually dipped into the Dewar flask with liquid nitrogen at the bottom keeping the film surface parallel to the appropriate temperature layer. In each sample position, the film electric resistance was recorded. Prior to annealing, the Fe–Al–Mn film has the resistance temperature coefficient (RTC) near to zero, that is typical of structural systems with films with different RTC sign. After annealing, the film has positive RTC. The temperature dependence of the film electric resistance after 90 minutes annealing at 573 K is shown in Fig. 2a.

A typical feature of the dependence is the presence of a nonlinear part in the temperature range 173–100 K. As shown in [4], such dependence is typical of metal transition from paramagnetic state to ferromagnetic one as the temperature drops below Curie point. As follows from those results, the $R(T)$ dependence is typical of metallic conductivity. The phase transition points determined from $dR(T)/dT$ derivative (Fig. 2b) show that the system has a complex phase composition. The peak at $T = 165\ K = T_C$ corresponds to the Curie point of Fe_2AlMn alloy [3]. This evidences the formation of the Heusler Fe_2AlMn phase due to diffusive annealing of vacuum-deposited films of each individual component. The nature of peak at $T = 129$ K can be associated with the appearance of second phase. As shown in [5], in a system consisting of two phases (paramagnetic and ferromagnetic) having semiconductor and metal conductivity character, espectively, the temperature dependence of resistance has a two-peak character.

A longer diffusive annealing (1 and 1.5 h) results in the further ordering of the film structure. At the same time, the magnetoresistance decreases down to zero (Fig. 3). This is due to homogenization of heterogeneous structure typical of Heusler alloys. It is possible that intermetallic phases have time to be formed during the long-term diffusive annealing. An evidence to this may be the resistivity increase above 650 K. This may result in formation of a homogeneous intermetallic structure. In spintronic systems, the best results are observed when using heterogeneous, in particular, granular structures. It is possible to avoid formation of the intermetallides during interpenetration of the film components by a short-term (10^{-3} to 10^{-8} s) laser annealing. It was found [6] that such pulse action results in mass transfer processes in the solid state with the mass transfer coefficient much exceeding the diffusion coefficient in the liquid metal. It should be noted that under pulsed laser action on the film deposited onto a ceramic substrate, it is possible to form a metal-ceramic film.

Thus, the results obtained in this work show a principal preparation possibility of Heusler type heterogeneous film structures using thermally activated mass transfer.

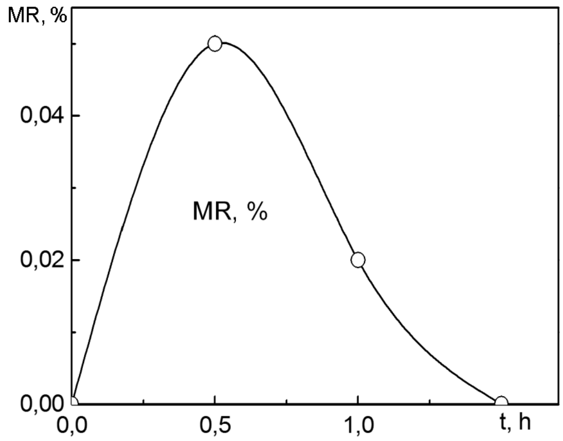

Fig. 3. Magnetoresistance (MR) of Fe–Al–Mn film vs annealing time ($T = 573$ K). Initial film thickness 400 nm.

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Отримання гетерогенних плівкових структур термічно активованим масоперенесенням

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Досліджено електричні та магнітні характеристики плівкових гетерогенних структур Cr(25 нм)/(Al + Mn) (50 нм)/Cr (25 нм) та Fe (100 нм)/(Al + Mn) (200 нм)/Fe (100 нм), отриманих за допомогою термічно активованого масоперенесення. Залежність $dR(T)/dT$ показала складний фазовий склад плівки після відпалу. Характерний пік при температурі 165 K відповідає T_C (точці Кюрі) сплаву Fe_2AlMn. Отримані результати свідчать про принципову можливість створення плівкових гетерогенних структур гейслерівського типу за допомогою термічно активованого масоперенесення.