Diffuse "amorphous phase 1 — amorphous phase 2" phase transition in the amorphous alloy Co–Ni–Fe–B–Si

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It is shown that under certain conditions relaxation annealing of the amorphous alloys Co–Ni–Fe–B–Si leads to a diffuse "amorphous phase 1 — amorphous phase 2" phase transition at the structural relaxation stage. © *1995 American Institute of Physics.*

Three types of structural changes caused by precrystallization annealing of amorphous alloys are associated with structural relaxation in these materials: decrease (redistribution) of the free volume during annealing, appearance of geometric (topological) short-range order, and establishment of chemical (composition) short-range order. A special case of structural changes of the latter type are stratification processes, which are recorded by direct and indirect methods, in amorphous alloys (see, for example, Refs. 1 and 2). In the present paper we show that an "amorphous phase 1 — amorphous phase 2" phase transition into the precrystallization temperature range is possible; i.e., another type of structural changes in amorphous alloys at the structural relaxation stage was discovered.

We investigated amorphous alloys of the system $Co_{58}Ni_{10}Fe_5B_{16}Si_{11}$ (CoNiFeBSi below), prepared by the plasma-deposition method in the form of up to 500-μm-thick coatings. The structural state of the coatings was monitored by x-ray crystallographic analysis (DRON-3), electron diffraction, DTA analysis, and magnetostructural methods.[3,4] (The relation of the morphological and structural features of the amorphous coatings to the main parameters of the deposition regimes is described in detail in Ref. 5. Here we note that the diffraction patterns of the amorphous coatings as well as characteristics such as the heat of crystallization (ΔQ) and temperature of crystallization (T_{cr}), saturation magnetization (M_S), spin-wave stiffness coefficient (D), and Curie temperature (T_C) are identical to the same characteristics of amorphous CoNiFeBSi foils fabricated at the Institute of Precision Alloys, Central Scientific-Research Institute of Ferrous Metallurgy, by the method of spinning of the melt.)

Figure 1 shows the thermomagnetic curves $M_s(T)$ of rapid heating of an amorphous CoNiFeBSi coating up to a temperature of 700°C and rapid cooling (30°C/min) of the amorphous-crystalline mixture formed. The temperature T_c of the amorphous coating and a manifestation of the crystallization temperature of the coating ($T_{\mathrm{cr}} \sim 540°\mathrm{C}$) can be seen in the heating curve — the increase in M_s at temperatures $T > T_{\mathrm{cr}}$ is due to the formation of a crystalline phase with $T_c > T_{\mathrm{cr}}$. The temperatures T_c of three ferromagnetic phases — amorphous and two crystalline phases comprising a dispersed mixture — are manifested on the cooling curve. One can see that when the cooling curve of the mixture is

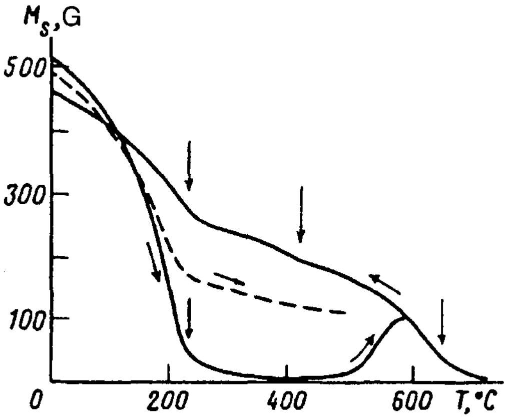

FIG. 1. Thermomagnetic curves (heating and cooling) of amorphous CoNiFeBSi coatings. The Curie temperatures of the amorphous and crystalline phases are marked. Dashed curve — Thermomagnetic heating curve of the amorphous alloy coating CoNiFeBSi synthesized at a substrate temperature of 300°C.

decomposed into partial curves $M_s(T)=\Sigma p_i M_{is}(T)$, where p_i is the volume fraction of the ith phase, the crystalline phase with $T_c \approx 620°C$ makes the main contribution to $M_s(T)$. It is well known that T_c of ferromagnetic amorphous alloys and their crystalline analogs differ by not more than 10–20% (see, for example, Ref. 6). Therefore, when the phase with $T_c \approx 620°C$ is obtained in the amorphous state, the difference in the temperatures T_c of this amorphous phase (2) and the initial amorphous phase (1) will be large. This is the basis of the method employed in the present work to record the phase transition "amorphous 1 — amorphous phase 2."

The appearance of dispersed inclusions of phase 2 in phase 1 was stimulated by varying the conditions of the deposition process. It was found that this is facilitated by heating of the base (on which the coating is deposited) up to temperatures from 180 to 380°C. In this case, an amorphous phase different from the initial phase is formed in the amorphous coating (see Fig. 1). (We note that ΔQ, T_{cr}, M_s, and D, which characterize the heterophase system formed, remain constant.) The transformation of the phase 1 into phase 2 is stimulated by relaxation annealing (annealing time 30 min).

The results of the thermomagnetic analysis of the annealed coatings are shown in Fig. 2. This figure also shows the temperatures T_{ic} of the phases formed versus the annealing temperature (T_{an}), as well as the partial contributions of the phases $y_i=p_i M_{is}/M_s$ to the curve $M_s(T)$ of the amorphous coatings. One can see that the initial amorphous alloy CoNiFeBSi can be characterized as a heterophase system in which phase 1 with $T_c \approx 260°C$ comprises 90% of the volume and the phase 2 with $T_c \approx 530°C$ comprises 10% (the fact that the temperature T_c of phase 1 is different from that of phase 2 indicates that the chemical short-range order in these phases is different.) This ratio of the volume fractions of the phases remains right up to annealing temperatures $\sim 250°C$. At $T_{an} \sim 270°C$ a ferromagnetic phase 3 with $T_c \approx 330-360°C$ arises; this phase remains an auxilliary phase ($y_3 \sim 15 \pm 20\%$) over the entire range of annealing temperatures. The behavior of the volume fractions y_1 and y_2 is interesting. One can see

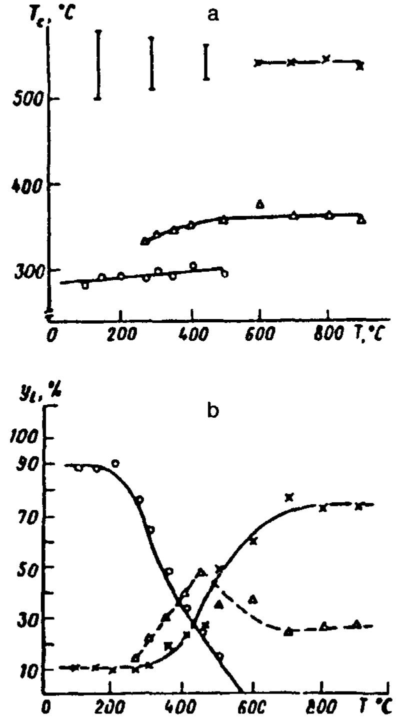

FIG. 2. Temperatures T_c (a) of individual magnetic phases and their volume fractions y_i (b) plotted as a function of the annealing temperature of the amorphous coating (annealing time 30 min). ○ — Amorphous phase 1; × — amorphous and crystalline phase 2; Δ — amorphous and crystalline phase 3.

(Fig. 2b) that as the annealing temperature increases, y_1 decreases and y_2 increases so that in the region $400<T_{an}<450°\mathrm{C}$ $y_1 \approx y_2$ and $T_{an} \sim 500°\mathrm{C}$ (which at 40°C is lower than T_{cr}) y_2 is much larger than y_1. Increasing T_{an} above 500°C at first causes the amorphous phase 1 to vanish and only then — at $T_{an} \sim 530°\mathrm{C}$ — results in crystallization (the structural state of the coatings was checked with x-rays). This behavior of the functions $y_1(T_{an})$ and $y_2(T_{an})$ indicates that the phase transition of phase 1 into phase 2, which occurs in the precrystallization temperature range, is a diffuse phase transition.

We note that the individual phases comprising the heterophase system of the amorphous CoNiFeBSi coating have one feature in common — they are ferromagnetic. Consequently , the strength of the local anisotropy field H_{ia} of the individual phases and the size r_{ia} of the orientational clusters comprising these magnetic phases are determined here from a special analysis of the magnetization curves $M(H)$ prior to saturation (method of correlation magnetometry,[3] adapted in Ref. 7 to heterophase systems). The quantities H_a and r_a are determined by the submicrostructural characteristics of the material (for example, H_a contains a contribution from internal stresses). Figure 3 shows the functions $H_{ia}(T_{an})$ and $r_{ia}(T_{an})$, which characterize the changes in the microstructure of the coating under the action of relaxational annealing. One can see that the

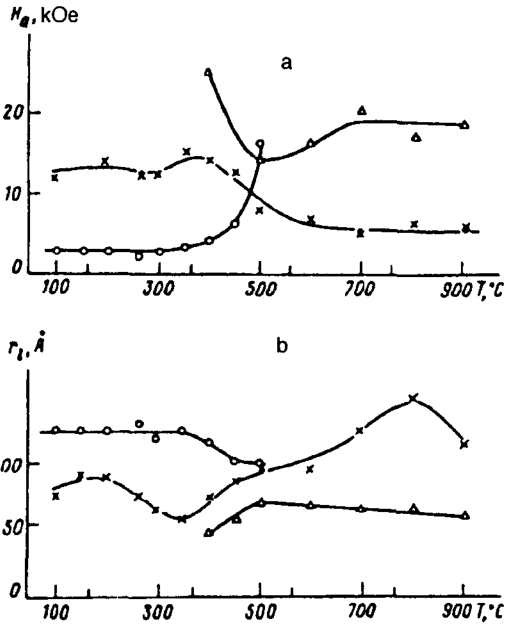

FIG. 3. Local anisotropy fields H_{ia} of individual magnetic phases (a) and their correlation radii r_{ia} (b) (same notation as in Fig. 2).

separation of the amorphous alloy and the diffuse phase transition are shown on these curves. The results obtained by us are physically reasonable: The main magnetic phase is characterized by a larger value of r_a and lower internal strain. In the auxiliary phases the internal strain is higher and the size r_{ia} of an orientational cluster is small.

In summary, in the present work we have shown that relaxational annealing of the amorphous alloy $Co_{58}Ni_{10}Fe_5B_{16}Si_{11}$ leads, under certain conditions, to a diffuse phase transition "amorphous phase 1 — amorphous phase 2" in this alloy in the precrystallization temperature range.

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