



Phase formation sequence, magnetic and structural development during solid-state reactions in 72Pt/28fcc-Co (001) thin films



V.G. Myagkov^{a,*}, L.E. Bykova^a, V.S. Zhigalov^a, A.A. Matsynin^a, D.A. Velikanov^a, G.N. Bondarenko^b

^a Kirensky Institute of Physics, Russian Academy of Sciences, Siberian Branch, Krasnoyarsk, 660036, Russia

^b Institute of Chemistry and Chemical Technology, Russian Academy of Sciences, Siberian Branch, Krasnoyarsk, 660049, Russia

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ABSTRACT

The phase formation sequence during the thermally induced solid-state reaction between polycrystalline Pt and epitaxial fcc-Co (001) films in the Pt/fcc-Co(001) bilayers are systematically examined using X-ray diffraction and magnetic measurements. The films have nominal atomic ratio Co:Pt = 28:72 and total thickness 300–400 nm. Annealing to the temperature of 375 °C does not change the structural and magnetic properties of the films; this is indicative of the absence of considerable mixing at the Co/Pt interface. With the subsequent increase of the annealing temperature, the phase formation in the Pt/fcc-Co(001) bilayers has been found to have two temperature (375 °C–575 °C and 575 °C–825 °C) intervals. Solid-state reaction between Pt and Co starts above 375 °C, and nanoclusters containing the ordered L₁₀ phase epitaxially intergrow with the disordered A1 phase of the composition CoPt₃ form and exist in the first temperature interval. The distinctive feature of the first interval is the formation of in-plane rotatable magnetic anisotropy. In the second temperature interval, the (L₁₀ + A1) two-phase mixture grows into the ordered L₁₂-CoPt₃ phase leading to the disappearance of rotatable anisotropy. Possible origin of the rotatable magnetic anisotropy is discussed. The first magnetocrystalline anisotropy constant of L₁₂-CoPt₃ has the maximum value of $-5.0 \cdot 10^5 \text{ egr/cm}^3$ and order parameter 0.55 at 675 °C. A careful analysis of thin film solid-state reactions implies the existence of low-temperature transformation (~375 °C) on the Pt-rich side of the Co-Pt system.

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1. Introduction

The Pt-rich Co_{1-x}Pt_x ($x > 0.7$), including CoPt₃ alloy films, has attracted a great deal of attention because of strong perpendicular magnetic anisotropy (PMA), which is important for a large number of practical applications. However, the origin of PMA in these films is not properly understood because it is not expected either in disordered A1- CoPt₃ or ordered L₁₂-CoPt₃ phases. Basic models developed to explain the PMA in these films are analogous to the multilayer case and are attributed to the formation of preferentially oriented to the film plane of the two-dimensional fcc Co nanoclusters within the Pt-rich matrix [1–10].

It is generally known that PMA is uniaxial anisotropy. Recently, high rotatable magnetic anisotropy (RMA), overcoming the demagnetization anisotropy energy, was obtained in epitaxial

L₁₀CoPt(111) films after the solid-state reaction in Co/Pt(111) bilayers [11]. The key feature of RMA is the rotation of the easy anisotropy axis behind the rotating magnetic field. In L₁₀CoPt(111) samples in magnetic fields above the coercive force, the easy axis can be oriented in any spatial (in-plane and out-of-plane) direction. It was concluded that the high RMA can be one of PMA sources in Co_xPt_{1-x} films [11]. Despite a large number of papers explaining the origin of the PMA in Co_xPt_{1-x} thin films [1–10], RMA in Co_xPt_{1-x} thin films is described in two experimental studies only. Park et al. reported Co_{0.47}Pt_{0.53} alloy films in which the location of the easy axis is not clear for the samples annealed above 500 °C [7]. Unfortunately, this fact is not interpreted correctly as the out-plane RMA phenomena. The small in-plane RMA was also observed in Co_xPt_{1-x} (Pt ~ 25 at%) films [12]. Magnetic characteristics of these films strongly depend upon fabrication methods and thermal treatment and other parameters, including the nature of the substrate, deposition temperature and annealing temperatures. The conventional methods of Co_xPt_{1-x} film preparation on a substrate at

* Corresponding author.

E-mail address: miagkov@iph.krasn.ru (V.G. Myagkov).

various growth temperatures reported in the literature are sequential sputtering deposition and electron beam co-evaporation from Co and Pt sources. A large number of studies have addressed the phase formation between elemental Pt and Co layers in bilayers and multilayers [13–25]. However, the conditions of the phase formation at the Co/Pt interface currently remain poorly analyzed.

In this work, we describe the solid-state reaction between polycrystalline Pt with polycrystalline Co and epitaxial fcc-Co(001) films. The final goal is to reveal a significant correlation between phase formation sequence and structural and magnetic transformations forming in the 72Pt/28fcc-Co bilayers after thermal annealing up to 825 °C. In addition, in numerous studies the origin of the PMA and the contribution of RMA to PMA remain unnoticed.

2. Experiment

The epitaxial Pt/fcc-Co(001) bilayers and polycrystalline Pt/Co bilayers were prepared by subsequent deposition of Co and Pt films on a freshly cleaved (001) surface of the MgO and glass substrate. Previously, the substrates were degassed at 350 °C for 1 h. The Co layer on the MgO(001) substrate was deposited at the pressure of 10^{-6} Torr and the temperature of 250 °C; this oriented the growth of fcc-Co(001) layer on MgO(001) with epitaxial relationship fcc-Co(001)[100] || MgO(001)[100] [26]. The fcc-Co(001)/MgO(001) films had a fourfold magnetic anisotropy with the constant K_1 coinciding with the first magnetocrystalline anisotropy constant of cubic cobalt fcc-Co ($K_1^{\text{fcc-Co}} = -(5.5-6.5) \cdot 10^5$ erg/cm³) [26]. The $K_1^{\text{fcc-Co}}$ is negative because the easy magnetization axes of the fcc-Co(001) film coincided with the [110] and [1–10] directions, which are the projections <111> on the (001) plane of the MgO substrate. The top Pt layer was deposited at room temperature to prevent reaction between and Co during the deposition. The thickness of the reacting Co and Pt layers, determined by the X-ray spectral fluorescent method, was ~80 and ~320 nm, respectively. This thickness ratio is close to the stoichiometry for CoPt₃. The mean chemical composition determined by EDS microanalysis was Co₂₈Pt₇₂. X-ray pattern did not contain Pt reflections; this means that the nanocrystalline Pt layer grows on the fcc-Co(001) (Fig. 1a). However, polycrystalline Pt layer grown on the polycrystalline Co film contained (111), (200), (220) peaks (Fig. 5a). The most plausible hypothesis of the essentially different Pt growth is that heat conductivity of MgO is about two orders of magnitude higher than of the glass. Therefore, deposited Pt atoms on Co/MgO are subject to a high undercooling rate, which it is considerably higher than on Co/glass substrate. As a result the nanocrystalline and polycrystalline structures are formed on MgO and glass substrates, respectively. As is known, nanocrystalline structures are frequently observed in metallic glasses under a deep undercooling. The deposition of the Pt layer did not change magnetic properties of the fcc-Co(001) and Co films. These data provide clear evidence that the initial samples are Pt/fcc-Co(001) and Pt/Co polycrystalline bilayers.

Initial Pt/fcc-Co(001) samples were annealed for 1 h from 25 °C to 825 °C with a step of 50 °C. The formed phases were identified with DRON-4-07 diffractometer (CuK α radiation). The epitaxial relationships between MgO(001), reacting Pt, Co films, and the Co_xPt_{1-x} layer formed in the reaction products were X-ray studied with PANalytical X'Pert PRO diffractometer with a PIXcel detector. The CuK α radiation monochromatized by a secondary graphite monochromator was used in the instrument. The saturation magnetization M_S and the coercivity H_C were measured with a vibration magnetometer in magnetic fields up to 22 kOe. Torque curves were measured on a torque magnetometer with a sensitivity of $3.76 \cdot 10^{-9}$ Nm and a maximum magnetic field of 10 kOe. All measurements were performed at room temperature.

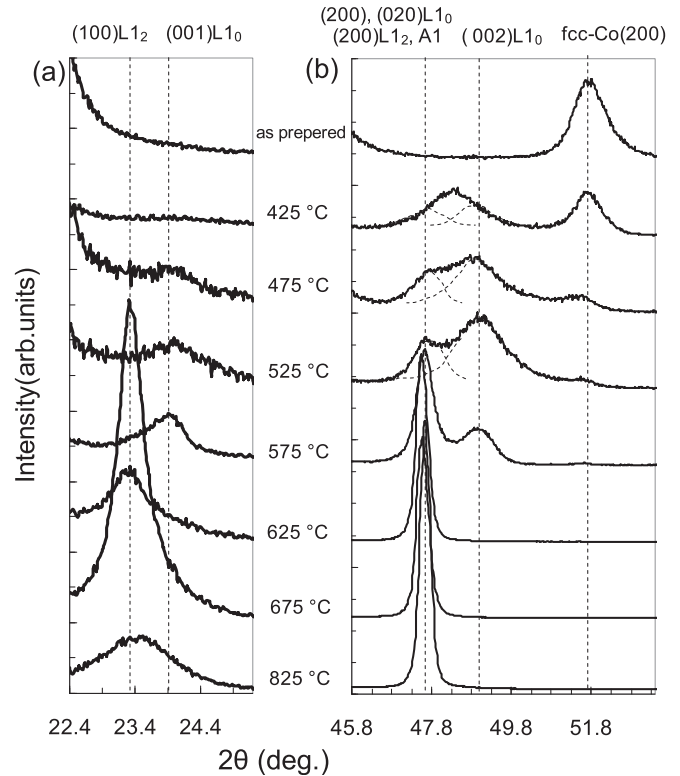


Fig. 1. Symmetrical X-ray scans through the (001)L10 and (001) L12 (a), the (002)L10, (200)+(020)L10, (200)A1, (002)L12 and (002)Co (b), reflections of epitaxial 72Pt/28Co(001) thin film before and after annealing (c). The annealing temperature changed from 375 °C to 825 °C.

3. Results

3.1. Phase transformations in 72Pt/28Co(001) thin films at increasing annealing temperature

3.1.1. X-ray studies

Fig. 1 shows XRD profiles of the as-deposited film and after annealing at temperatures from 375 °C to 825 °C for 1 h. Up to ~375 °C, the X-ray patterns of the Pt/fcc-Co(001) bilayers did not change; this is indicative of the absence of mixing and reaction between the Pt and Co layers. Analysis of diffraction data shows two (375 °C - 625 °C and 625 °C - 825 °C) temperature intervals. In the first temperature intervals after annealing at 375 °C, strong (200) fcc-Co peak began decreasing and weak (001) and {200} reflections from the ordered L10 and disordered A1 phases appeared. The (001) reflection with an interplanar spacing $d_1 = 0.372$ nm appears at approximately 24° and it is an essential sign of the L10 phase only. It is important to note that the nonequilibrium process of mixing Co and Pt layers above 375 °C immediately results in the ordered L10 phase. Above 425 °C X-ray diffraction reflections appear with interplanar spacings $d_2 = 0.191$ nm and $d_3 = 0.186$ nm. We associate d_1 and d_3 peaks with the (001) and (002) reflections from the Z-variant of ordered L10 phase. The d_2 peak may belong to the (200) and (020) reflections from the in-plane X-, Y- variants of L10, respectively, and (200) of disordered A1 (Fig. 1b). These peaks progressively increase with annealing temperatures up to 575 °C, as a result of increasing volume fractions of L10 and A1 phases. After annealing at 575 °C intensity ratio $I(200)/I(002) \sim 3.2$, it is appreciably more than 2, which corresponds to the complete ordered L10 with equal the volume fractions of X-, Y-, Z- variants. This suggests that besides L10 a considerable proportion of the disordered A1 was

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