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Effect of low-energy ion bombardment during the sputtering on the crystal structure of FePt films

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The crystal structure and texture of FePt films have been studied. The film were grown on Si and Al_2O_3 substrates by RF magnetron sputtering using ion bombardment during the growth. The ion bombardment was done by applying an RF bias to the substrate. At room temperature of substrate, the effect of external magnetic field directed along the substrate surface during the film growth was studied. The films with axial (111) texture have been obtained at any substrate type. The substrate bias and magnetic field enhance the film crystallinity degree without change of texture type. The films with in-plane magnetization have been obtained at Al_2O_3 substrate temperature above 400°C and at substrate bias of 3 to 5 V but without use of magnetic field.

Исследованы кристаллическая структура и текстура пленок FePt. Пленки получены ВЧ магнетронным методом с использованием ионной бомбардировки в процессе роста на подложках Si и Al_2O_3. Ионная бомбардировка осуществлялась путем подачи на подложку ВЧ смещения. При комнатной температуре подложки исследовалось влияние магнитного поля, приложенного вдоль поверхности подложки в процессе нанесения пленок. Получены пленки с аксиальной текстурой (111) независимо от типа подложки. Смещение на подложке и магнитное поле усиливают степень кристалличности пленок без изменения типа текстуры. Пленки с намагниченностью, лежащей в плоскости подложки, получены на подложках Al_2O_3 при температуре выше 400°C с использованием смещения на подложке 3-5 В, но без использования магнитного поля.

At present, commercial application of high coercivity media for magnetic recording is complicated by the absence of recording heads capable to magnetize alternatively such high-coercive media. One of ways of writing head manufacturing is proposed in [1]. Basic element of head is the pair of giant anisotropy magnets (plates) put together along a direction of magnetization in antiparallel. At the edge of opposite magnetic poles the strong stray field exists. Instead of two plates it is better to use layers which are made by thin-film technology.

The purpose of the work presented is to estimate an opportunity of sputtering a FePt film in such a way, that the magnetization direction was lying in the plane of the substrate surface.

FePt compound is a hard magnetic material with high magnetic anisotropy [2]. Its magnetic structure is completely determined by crystal structure. The crystal structure is ordered as $L1_0$ type, similar to the crystal structure of CuAu alloy. At the temperature T above the critical value T_k a FePt monocrystal has face-centered cubic (fcc) crystal lattice. At $T < T_k$ the face-centered tetragonal (fct) lattice becomes equilibrium. During the transition fcc → fct any of cubic axes [100], [010] or [001] can become a tetragonal axis. Thus, three orientations of **c** axes are possible. We shall designate them **c1**, **c2**, **c3**, correspondently. Structural transformation fcc → fct is developing as follows. In initial monocrystal of an fcc phase the nucleating centers of the fct phase occur, which have a form of fine microplates. Around a microplate the elastic stresses arise due to the difference between the parameters of fcc and fct lattices. In order to diminish elastic energy, the growth of fct phase volume is developing by the formation on the initial microplate (whose tetragonal axis is **c1**, for example) of the following plate, whose tetragonal axis **c2** forms with an axis **c1** a corner of 90°. Such pair of microcrystallites is a twin structure. The next layer is a plate similar to the first one, the next is similar to the second one, etc. As a result, a large polytwin plate is being formed. There is a correspondence between the complicated structural hierarchy and the hierarchy of magnetic domain system, which is no less complicated. Every microplate is a magnetic microdomain. The twin crystal border between neighbouring plates with axes **c1** and **c2** (or **c1** and **c3**, or **c2** and **c3**) is simultaneously a magnetic 90° wall. But, unlike usual 90° walls in homogeneous crystals, this wall cannot be displaced in any magnetic fields. It is a "frozen" wall. These microdomains form a macrodomain magnetic structure. Each macrodomain covers tens, hundreds and thousands of microdomains. Macrodomain structure is also called a cooperative domain structure (CDS). Macrodomains are divided by 180° domain walls, as in uniaxial magnetic crystals. These walls are mobile; they can be displaced under the action of a magnetic field. The "frozen" and mobile walls set leads to originality of the CDS reorganization in a "polytwin" crystal under the action of a magnetic field.

Naturally the sputtering of FePt films should possess an additional specificity. In particular, the epitaxial growth on a substrate with square atomic structure of a surface and appropriate atomic conformity of a film can lead to the formation of a structure, whose **c** axis is perpendicular to the substrate surface. However, with a growth of thickness such a structure will become less and less stable, because the twin structure is more favourable energetically. Authors of work [2] assert, that a twinning plane for FePt belongs to {101} family of planes. It means, that the growth of thickness of the films with (001) orientation should lead to the formation of the grains, whose **c** axes are almost parallel to the surface.

On an amorphous substrate in most cases also films with orientation (001) are being formed, but in this case they will be axial, in a greater or lesser extent, and the texture axis [001] will be perpendicular to the substrate surface.

An application of magnetic field during the process of films sputtering may be quite effective for producing of the films with perpendicular anisotropy, about which it was spoken above. We can suppose also, that in order to obtain microdomains with smaller sizes an application of a variable magnetic field should be used, with a vector of intensity oscillating along a normal to the substrate surface.

The formation of the FePt films with the residual magnetization oriented along the substrate surface, in our opinion, is possible only on substrates with asymmetrical surface atomic structure. In this case various orientations of the twin structures are possible. In particular, **c** axes of the microplates can be placed along the substrate plane and also under some angle to it. The structure with various volume fractions of microplates can be realized. Generally, the structure will be determined by the substrate used and the technological features of sputtering process. The using of the longitudinal magnetic field during the sputtering will not be effective, as it will lead to the formation of a great volume of the film with **c** axis, oriented along the field, while the producing of the films with good magnetic characteristics requires the presence of equal quantities of volumes with perpendicular **c** axes. Besides, it is necessary to know precisely the required orientation of the magnetic field concerning the crystallographic direction of the substrate.

In the presented work the Si substrates with the orientations (111) and (001) and Al_2O_3 substrates with the orientation $(01\bar{1}2)$ were used. The FePt films were sputtered by the radio-frequency (RF) magnetron method with using of RF bias on the substrate. The target was a Pt disk with Fe segments. The Fe segments on the Pt disk were located with central symmetry. The composition of the target was variated by changing the number of the Fe segments. As a characteristic of the target composition, the proportion of areas S_{Fe}/S_{Pt} was used. Besides, the composition of films was changing due to the variation of the target power and the bias value, as will be shown below. All the films, presented in this work, have a thickness of 0.2 μm. The technology of sputtering is presented at length in [3, 4].

The distinctive feature of the presented work is the using of the magnetic field (approximately 0.15 T) during the films growth process. The field was directed along the substrate surface. Unfortunately, the magnets used were NdFeB rods with low value of the Curie temperature (~100°C), therefore the influence of a magnetic field has been investigated only in such a case, when the substrateholder is cooled by water.

The chemical composition of the films was monitored by means of Auge-spectrometry. The method of X-ray diffraction (XRD, copper target, pyrolitic graphite monochromator) was used for the investigation of the crystallographic structure of the films. The analysis of the films texture was realized by using the technique of the polar figures. The STM images were obtained on SMM 2000 microscope.

a. Growth of FePt on cooled substates. The investigation of the FePt films with Si substrates showed that the following parameters influence on the crystal structure of the films: the bias on a substrate, presence of the magnetic field and the target power (rate of sputtering). All the produced films possessed the axial structure (111), expressed in a greater or lesser extent. Therefore, if it is not mentioned especially, we shall use the concept of the crystallinity degree, which in the first approximation corresponds to the intensity of X-ray peaks (111) and (222), and we shall not mention that the structure is axial. It concerns the investigations of the films, produced on Si substrates. In this case even the using of the magnetic field during the sputtering did not lead to the formation of crystallographic anisotropy of the films in the plane of the substrate.

In the Fig. 1 the X-ray difractograms of the FePt films are presented, which show the influence of the substrate bias on the crystallinity degree in the presence and in the absence of the magnetic field. We can see that the increase of the substrate bias in both cases leads to the increase in the crystallinity degree. This influence is more considerable for the films, produced in the magnetic field.

Measurements of the chemical composition of two films, produced in the magnetic field, were carried out: at the "floating" potential of the substrate and at the offset of 10 V. In the first case the composition was $Fe_{0.41}Pt_{0.59}$, in the second — $Fe_{0.32}Pt_{0.68}$. The composition of the target was $S_{Fe}/S_{Pt} = 9/7$, the power on the target was 150 W. The "floating" potential of the substrate takes place in a case when the substrateholder is connected neither with the RF source, nor with the installation's case. Usually, due to the action of the target's discharge the substrateholder is being charged up to a small (5 V) positive potential concerning the installation's case.

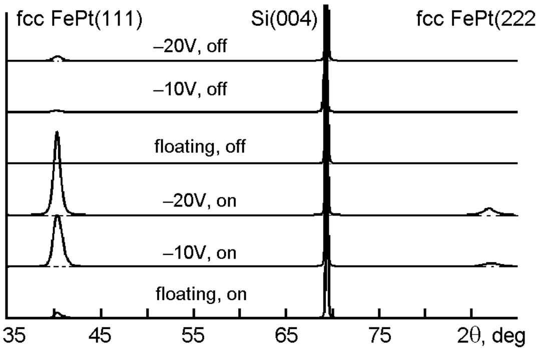

Fig. 1. X-ray difractograms of the FePt films, produced at various values of the substrate bias ("floating", −10 V, −20 V) in the presence (on) and in the absence (off) of the magnetic field. The power on the target is 300 W, the proportion of areas Fe/Pt is 1/1, the Ar pressure is 0.5 Pa. The chemical composition was measured for the sample "floating, off" ($Fe_{0.33}Pt_{0.67}$).

Analogical structural investigations were carried out for the films produced in the magnetic field with a target of the composition $S_{Fe}/S_{Pt} = 5/3$, the target power was 500 W. Besides it was revealed, that with increase of the substrate bias the XRD peaks are being displaced to smaller angles. It shows that the parameters of the FePt crystal lattice were changing. Apparently, this change corresponds to the change of the films chemical composition due to the above-mentioned increase of the bias. Measurements of the chemical composition for these films were not carried out.

The Fig. 2 shows the X-ray difractograms of the FePt films produced at various values of the target power in the presence and in the absence of the magnetic field. Time of the sputtering was chosen in inverse proportion to the value of the power so that the films' thickness was identical. We can see that with the increase of the target power the crystallinity degree increases in the presence of the magnetic field. In the absence of the magnetic field there is no such effect.

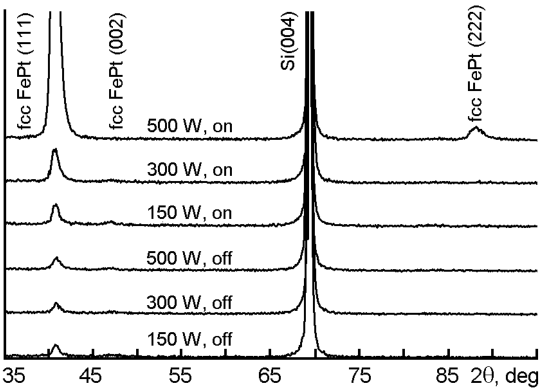

Fig. 2. X-ray difractograms of the FePt films, produced at various values of the target power (150 W, 300 W, 500 W) in the presence (on) and in the absence (off) of the magnetic field. The potential on the substrate is "floating", the Ar pressure is 0.5 Pa. The target composition is S_{Fe}/S_{Pt} = 5/3.

Thus, polycrystalline axial FePt films are obtained on the Si substrates. Parameters of the crystal lattice **c** lie on the surface of a cone whose axis is perpendicular to the substrate surface. The angle between **c** axes of the crystallites and the substrate surface is 36.2°.

b. Growth of FePt on heated substrates. The following results were obtained in the investigations of the crystal 01$\bar{1}$2 structure of the films produced on Al_2O_3 substrates, with the surface oriented as . The FePt films produced on the substrates cooled with water are similar to the films produced on the Si substrates. So we shall describe at length the results obtained for the samples on Al_2O_3 substrates, being heated during the sputtering. The magnetic field was not applied because of the above-mentioned reasons.

The Fig. 3 shows X-ray difractograms of the films produced on Al_2O_3 substrates with various substrate biases. The substrates were heated up to the temperature of 600°C. In the case of the "floating" potential on the substrate the film has polycrystalline structure. There are grains with orientation of (001), (100) (peak (200)), (110), (111).

The using of the substrate bias during the growth leads to the suppression of growth for all grains whose orientation is not (110). Such orientation corresponds to the placement of the crystal lattice parameter **c** in the substrate plane. Moreover, we can see that the using of the bias stimulates the growth of grains with (110) orientation just like it was in the case of grains with orientation (111) on Si, especially in the presence of the magnetic field. It should be mentioned, that it takes place on a heated substrate. In the case of the target composition S_{Fe}/S_{Pt} = 5/3, when the potential on the substrate is "floating", also only polycrystalline films were obtained. Thus, we can assert, that the basic parameter, which influences on the formation of a film with (110) orientation, is the substrate bias.

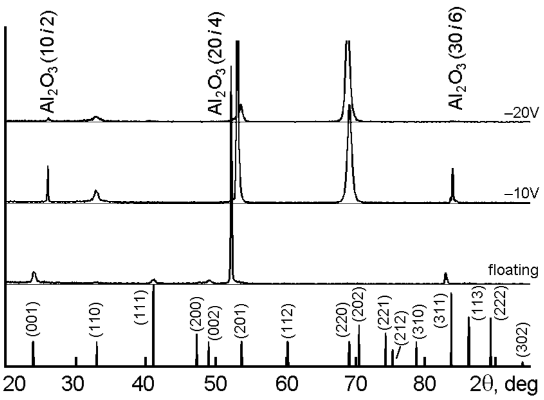

Fig. 3. X-ray difractograms of the films, produced on Al_2O_3 substrates at various values of bias ("floating", –10 V, –20 V). The temperature of the substrate is 600°C, the power on the target is 300 W, the chemical composition of the target is S_{Fe}/S_{Pt} = 9/7. The difractogram in the lowest part of the figure corresponds to the polycrystalline sample FePt (the card: PDF 26-1139).

Let's note one more important point. The lower part of the Fig. 3 shows a difractogram of a bulk polycrystalline FePt sample. The comparison of this spectrum with two difractogram for the samples produced with the substrate bias shows the discrepancy of intensities for the peaks of the first and the second order, (110) and (220). In the case of a crystal they must have approximately identical intensity, but in the films the intensity of (110) peak is much less than the intensity of (220) peak. This fact is due to the insufficient ordering of atoms in (110) plane of the films. In the ordered state, these planes should be completely (through one plane) filled by atoms either Fe, or Pt. We can see it in the Fig. 4.

The vertical planes, parallel to the cell parameter **c**, as well as parallel to the horizontal diagonal of a cell, pass either only through Fe atoms, or only through Pt atoms. It should be mentioned that in the case of fcc lattices with identical atoms (Pt, for example) the (110) XRD peak is forbidden, so this peak doesn't occur on the Pt difractogram. Just the presence of the al-

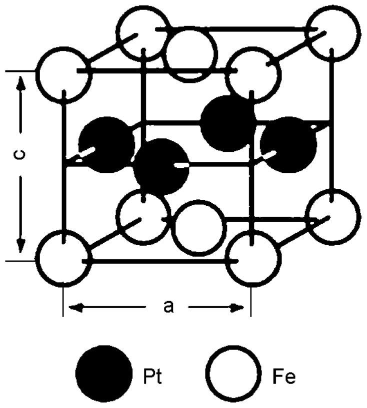

Fig. 4. The unit cell of FePt.

ternating planes (220), which contain various atoms, leads to the break of the condition of disappearance for (110) peak, due to the different diffract ability of Fe and Pt atoms. Therefore, though X-rays, diffracting on the lattice formed by (220) planes, come to the detector in antiphase, but their intensity is different, so the peak appears. But this peak can decrease or disappear even for FePt alloy, if the ordering shown on Fig. 4 will not occur — when (220) planes, which must contain only Fe atoms, contain also Pt atoms, and the reverse. Besides, the diminishing of the intensity of (110) peak can be in the case of non-strict alternating of Fe-planes and Pt-planes.

Thus, the ordering in the films produced with the substrate bias (their difractogram are presented in the Fig. 3) is not well enough. One more probable reason, in our case, is a deviation from the stoichiometric composition of FePt in the film. The best proportion of the peaks intensities $I_{(110)}/I_{(220)} = 8/11$ has been obtained in a film produced at the following conditions: the substrate bias was 0 V, the power on the target was 100 V, the temperature of the substrate was 650°C. In order to keep the potential of the substrate to be equal to the potential of the installation's case, the RF power applied to the substrateholder was 5 W.

The least proportion of the peaks intensities $I_{(110)}/I_{(220)} = 50/4000$ has been obtained for a film produced at the following conditions: the offset on the substrate was −5 V, the power on the target was 300 V, the temperature of the substrate was 400 °C. It was noticed, that when a good proportion $I_{(110)}/I_{(220)}$ is achieved, a small amount of grains with orientation (111) appears.

The Fig. 5 shows the difractograms of two FePt films produced at various values of the target power (100 W and 150 W). We can see that a film, produced at 150 W on the target, contains only grains with (110) orientation. Another film, produced at 100 W on the target, contains grains with two orientations: (111) and (110).

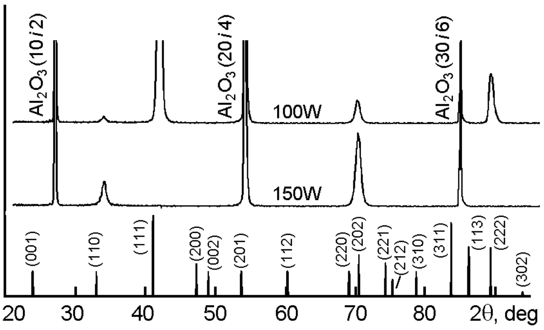

Fig. 5. X-ray difractograms of the films, produced at the following conditions: the composition of the target is $S_{Fe}/S_{Pt} = 5/3$, the substrate bias is −5 V, the temperature of the substrate is 650°C, the target power is 100 W and 150 W. On the top difractogram the intensity of (111) peak is 5000 (imp/s), the intensity of (222) peak is 400 (imp/s).

Further, the investigation of the texture of two films is presented. The difractogram of the samples are shown on the Fig. 5. The research was carried out by a method of construction of the polar figures. The polar figure (hkl) can be constructed as follows. The detector is mounted on an angle 2θ, which corresponds to a pick (hkl). The sample turns around the axis, which is perpendicular to the axis of the goniometer's rotation and is located in a plane of the sample, with an angle of inclination α. Then, while the detector is on, the sample turns 360° around its own normal to its surface. Then the curves, obtained at various values of α, can be reduced to a polar figure.

Thus, the polar figure (hkl) shows the angular distribution of normals to the investigated crystallographic planes (hkl). In the case of a monocrystal this distribution consists of points corresponding to each normal [hkl]. In the case, when there is a texture in the sample, we represent the density of normals as the gradation of grey color. Black color designates the maximal density of normals. The polar figures are constructed in a scale of a gnomostereographic projection (Woolf's grid). The deviation angle from the normal to the surface is laid from the center of a polar figure along radius. The point on the external circle has an inclination angle of 90°, i.e. the corresponding normal is lying in the plane of

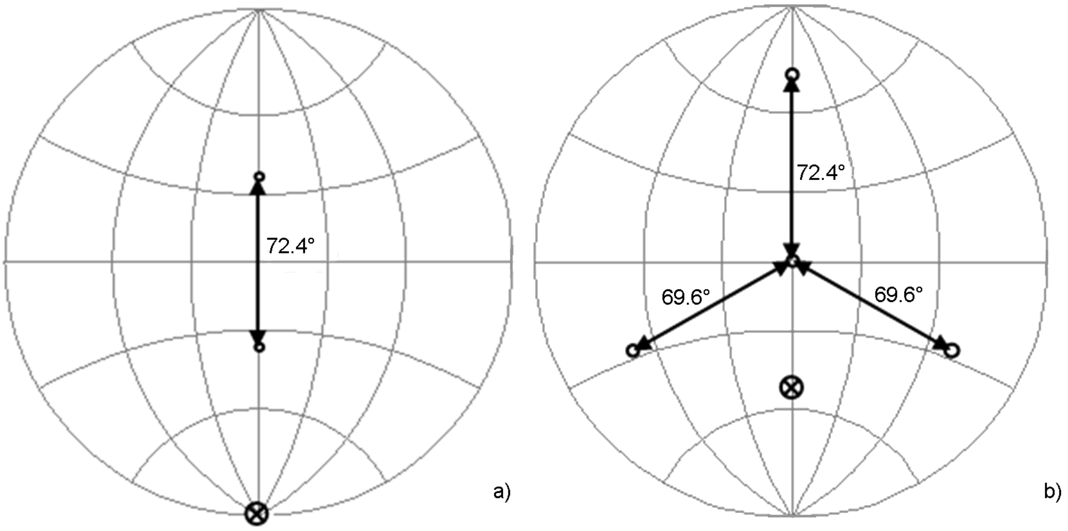

Fig. 6. Polar figures (111) for ideal single-phase FePt films with (110) orientation (a) and (111) orientation (b). The circles with crosses designate the pole, which will be visible on the polar figure (001).

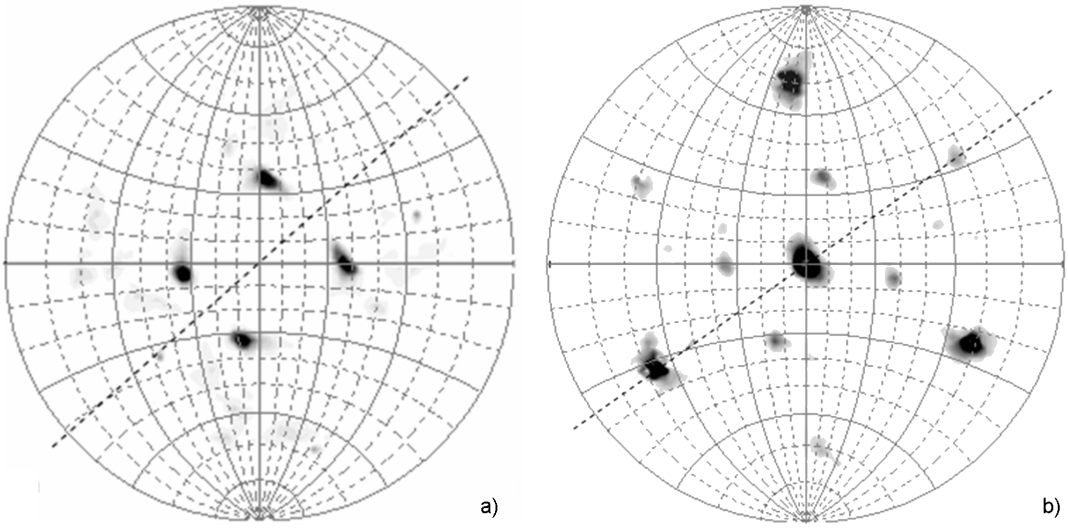

Fig. 7. Real polar figures (111) for the films produced at the power 150 W (a) and 100 W (b) on the target. Dashed lines designate the direction $[01\bar{1}1]$ in the plane of Al_2O_3 substrate (see Fig. 8).

the substrate. Along the circle the azimuthal angle is laid. In Fig. 6 the polar figures (111) for ideal single-phase FePt films are shown.

The Fig. 6a is a polar figure (111) for the film with (110) orientation, where the **c** axis is parallel to the vertical axis of the polar figure. The Fig. 6b is a polar figure (111) for the film with (111) orientation, where the **c** axis goes under an angle of 36.2° to the substrate surface. On both polar figures the position of the **c** axis is shown as a circle with a cross. In these places there will be a pole on a polar figure (001) or (00*l*).

The Fig. 7 shows the real polar figures of the films, whose difractograms are presented in the Fig. 5. The film with (110) orientation (the distance between poles is closer to the value of 72.4°, than to the value of 69.6°) (Fig. 7a) has approximately identical quantity of grains with almost perpendicular direction of **c** axes. An angle between them is a little less than 80°. If there was a twinning along (111) plane, which is perpendicular to the substrate surface, then the polar figure would be the same, but the angle between **c** axes would be equal to 74°.

The film with (111) basic orientation (Fig. 7b) has grains with two various directions of the lattice in the plane. One of these orientations corresponds to the central pole and to the intensive poles located under an

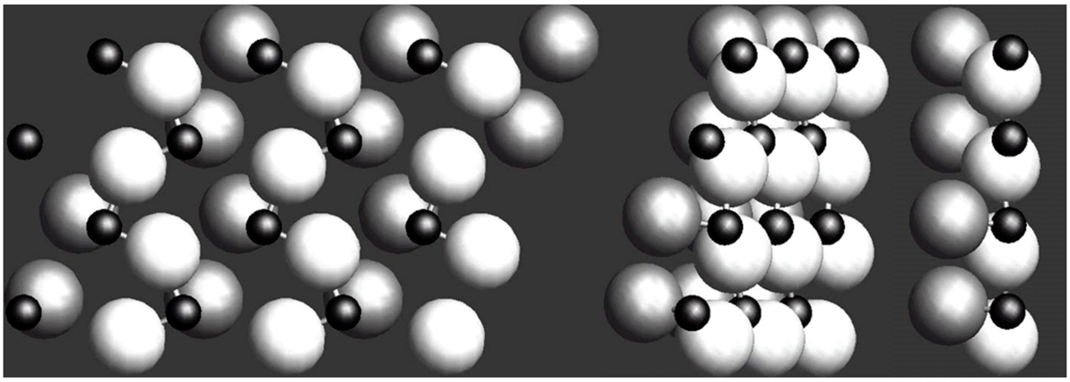

Fig. 8. Three views of the same fragment of Al_2O_3 surface — $(01\bar{1}2)$ plane. From left to right: top view, at an angle of 60°, and at an angle of 90°. The crystallographic direction $[11\bar{1}0]$ of Al_2O_3 is parallel to the horizontal of the left image; the crystallographic direction $[01\bar{1}1]$ is lying along the vertical of the figure. Small dark spheres are aluminium, greater light spheres are the oxygen, two layers of oxygen differ one from the other in their tone.

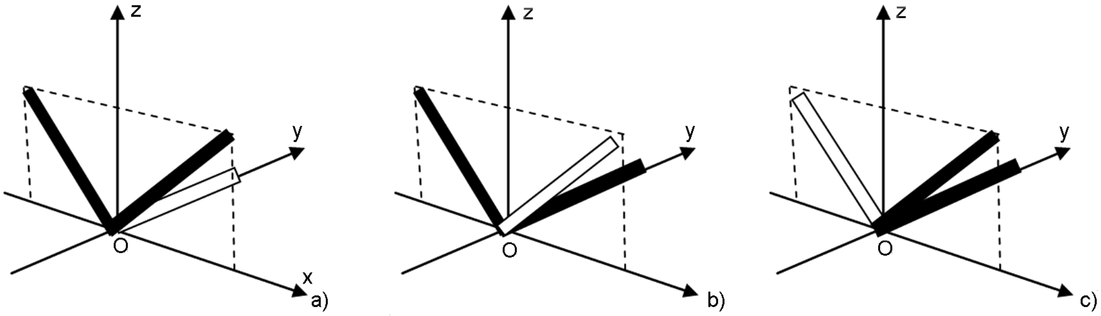

Fig. 9. The location of the elementary cell's edges in the cases: a) the single-phase film with (110) orientation, b) and c) the variants of twinning along (101) planes. The substrate plane coincides with xOy plane. Black rectangles in the figure designate the parameter **a** of the lattice, white rectangles designate the parameter **c**.

angle of 120° to each other. The **c** axis of these grains is under an angle of 36° to the substrate surface, and its projection, apparently, is lying along the direction depicted by a dashed line on the polar figure. The second orientation corresponds to the poles with weak intensity, which are mirror-symmetrical to the poles of the orientation already described concerning the direction which is perpendicular to the direction depicted by a dashed line. The **c** axis of such grains is also mirror-symmetrical. It is supposed, that the basic problem will be a twinning during the magnetic annealing. If twinning occurs just like it has been described in the beginning of the paper, along (101) plane, then the edges of an unit cell of the recrystallization twin, **a** and **c**, will be located, correspondently, just like **c** and **a** edges of the initial grain. It means that in the case of (110) orientation of the grain, the twin will have (101) orientation, i.e. the **c** axis of the twin will be located under an angle of 45° to the substrate and under an angle of 90° to **c** axis of the initial grain. In total, there are only two variants of a spatial location of **a** and **c** axis of the twin for one initial grain, if we are not to consider the distinctions in 2° (Fig. 9).

If the orientation of the initial grain is (111), the twinning will lead to the turn of **c** axis around the normal to any of (111) planes on an angle of 120°, including the normal to the substrate surface, because one of (111) planes coincides with it. Last case on a polar Fig. 6b looks as turn of poles around of the center of a polar figure on 120°. In case of (111) orientation there will be six different variants of twinning for one concrete grain.

The Fig. 10 shows STM images of a film with (110) orientation, used for estimation of the crystallites sizes. The first image (Fig. 10a) was made after the sputtering. At scanning STM images the influence of scanning parameters (voltage on a needle and tunnel current) on image structure was revealed. It has been assumed, that the reason of it can be influence of film‘s magnetic structure on the form of a tunnel current way. To confirm this assumption, the image of the same sample

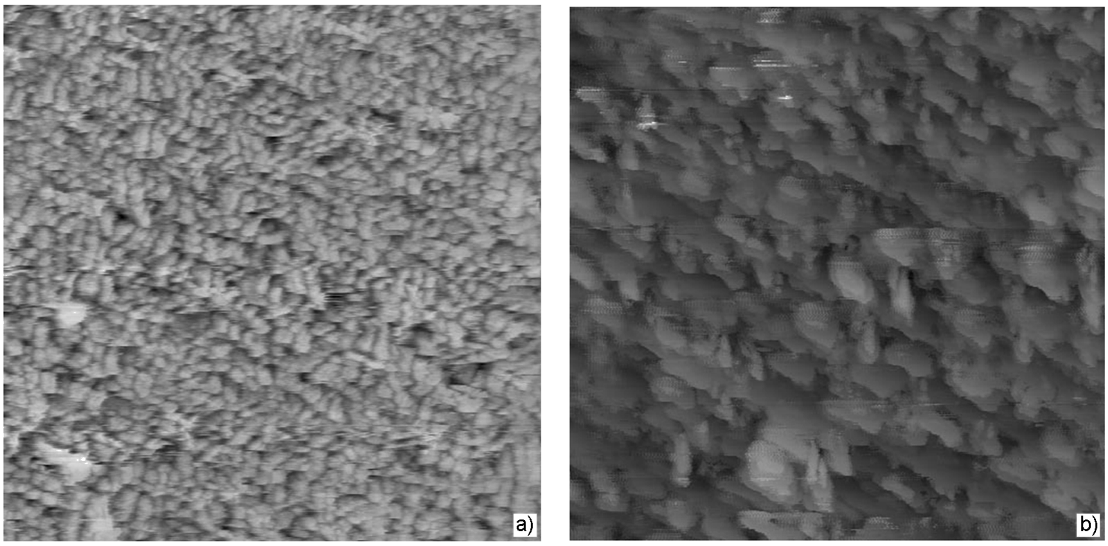

Fig. 10. STM images of a film with (110) orientation a) after sputtering and b) after magnetization along the vertical direction. The size of images is 1.2×1.2 μm^2.

has been received at the same scanning parameters after its magnetization outside of a microscope. Corresponding STM image is shown on Fig. 10b. The microscope has not been equipped by any magnetosensitive devices.

In summary we shall note, that the crystal structure and texture FePt films strongly depends on technological modes of their obtaining. The basic role in reception of in-plane orientation of **c** axis of crystal unit cell is played with two factors: the atomic order of a substrate surface and weak ionic bombardment of a film during its growth.

Reasonable magnetic characteristics FePt films are received after magnetic annealing. Annealing was carried out in other group. Predictably, magnetic characteristics of films have appeared are complex for interpretation. For these reasons results of researche of magnetic properties will be published later. Further the structure from two FePt films separated by non-magnetic layer will be investigated.

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Вплив бомбардування низькоенергетичними іонами під час розпилення на кристалічну структуру плівок FePt

В.В.Наумов, Е.І.Іл'яшенко

Досліджені кристалічна структура і текстура плівок FePt. Плівки одержані на підкладках кремнію та Al_2O_3 методом ВЧ магнетронного розпилення з використанням іонного бомбардування в процесі росту. Іонне бомбардування здійснювалося шляхом подавання на підкладку ВЧ зміщення. При кімнатній температурі підкладки досліджено вплив магнітного поля, прикладеного вздовж поверхні підкладки у процесі нанесення плівок. Одержано плівки з аксіальною текстурою (111) незалежно від типу підкладки. Зміщення на підкладці та магнітне поле збільшують ступінь кристалічності плівок без зміни типу текстури. Плівки з намагніченістю у площині одержано на підкладках Al_2O_3 при температурі вище 400°С з використанням зміщення на підкладці 3-5 В, але без використання магнітного поля.