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Studying the magnetoelastic effect in submicron Ni particles on lithium triborate single-crystal surface

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The influence of thermoinduced magnetoelastic effect on the coercivity field of submicron rectangular Ni particles with respect to orientation of their long side to axes of the crystalline substrate was studied. For this purpose, particles with size of $0.9 \times 0.3 \times 0.03 \,\mu$ m were formed on the single-crystalline lithium triborate (LiB₃O₅) surface at angles of 0, 20, 50, 65, and 90° relative to the *z*-axis of the single-crystal. It was experimentally shown that by changing the sample temperature in the range of $25-55^{\circ}$ C, it is possible to both decrease and increase the particle's coercivity field. The observed changes in the coercivity field are associated with the magnetoelastic anisotropy induced in the particles due to differences in the thermal expansion coefficients of the substrate along different crystalline axes and the angle between the particle's long side and the *z*-axis of the single-crystal.

Keywords: thermally induced magnetoelastic effect, submicron particles, switching field, magnetic force microscopy.

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1. Introduction

Currently, one of the problems of using magnetic media for recording and storage of information is two mutually exclusive requirements. On the one hand, a high coercive force of the information storage medium is required for increasing the noise immunity and storage time. On the other hand, a low coercive force of the medium is required for recording information for reducing the energy consumption of such devices. Several method have been proposed to solve this problem. In particular, it was proposed to reduce the coercive force by heating — this is the so-called method of Heat-Assisted Magnetic Recording — (HAMR) [1–3] and by using the magnetic elastic effect [4–6].

A bit of information is recorded by an external magnetic field at the time of heating of a separate ferromagnetic particle acting as a carrier of a bit of information in case of usage of HAMR. A focused pulsed laser radiation can be used for heating [1]. In this case, the coercive force of the particle is significantly reduced by heating, and information can be recorded at sufficiently small values of the external magnetic field. Information is stored and read at room temperature. The disadvantage of this method is the need to use high temperatures — close to or even above the Curie temperature of the particle [2].

The application of the magnetic elastic effect reduces the coercive force by creating induced magnetic elastic anisotropy in the particles. They can be formed on a substrate with an anisotropic coefficient of thermal expansion for this purpose, for example, polyvinylidenfluoride (PVDF) [4] or lithium triborate (LiB₃O₅) [6]. It is also possible to use substrates that experience a phase transition when heated/cooled with the change of the dimensions of the lattice cell in one direction, as in the case of substrates with an anisotropic coefficient of thermal expansion. Similar studies were performed on a vanadium oxide substrate [7]. Other methods of creation of a magnetic elastic effect in a sample are provided in [8,9].

The simultaneous use of heating and the magnetic elastic effect may be a promising solution for reducing the coercive force when recording information. From this point of view, ferromagnetic particles are of the greatest interest, in which the state of homogeneous or quasi-homogeneous magnetization turns at 180° and persists after switching off the external field and lowering the temperature of the sample to the initial level (i.e., the temperature of information storage). "Quasi-homogeneous magnetization" here means such a state of the magnetic subsystem of an object with dimensions noticeably larger than the single domain radius, in which, a multi-domain state is not formed due to the anisotropy of the shape. Usually, the planar particles having the shape of a strongly elongated ellipse, triangular and quadrangular particles with varying degrees of concavity of the sides have the quasi-homogeneous magnetization structure [10,11].

We have previously conducted studies with the same Ni particles on a LiB_3O_5 substrate [6]. However, only one direction was considered, relative to which uniaxial mechanical stresses of compression and tension were created in the particles. The external magnetic field was always directed along the long side of the particle. It was found that the magnitude of the switching field of such particles can be reduced by 1.57 times by increasing the sample temperature

by only 15°C. At the same time, the question remains open: how will the switching field of a particle change if a uniaxial mechanical stress and an external magnetic field are induced at different angles relative to its sides. It is well known that the coercive force of a particle depends on the direction of application of the external magnetic field [12] and on the direction of induction of the magnetic elastic anisotropy [6,9,13]. This paper studied the effect of the thermally induced magnetic elastic effect on the switching field of planar rectangular particles depending on the orientation of these particles relative to the *z*-axis of a lithium triborate substrate (LiB₃O₅).

2. Methods of sample preparation and measurement

Ni particles for measurements were formed on the surface of a single-crystal lithium triborate (LiB₃O₅, hereinafter LBO) produced by HG Optronics. The technique described in detail in Ref. [6] was used for this purpose. The plane for particle formation was formed by the crystal axes x and z. According to the manufacturer, the coefficients of thermal expansion of LBO along the x-axis were $\alpha_x = 10.8 \cdot 10^{-5} \circ \text{C}^{-1}$, along the z-axis $\alpha_z = 3.4 \cdot 10^{-5} \circ \text{C}^{-1}$. A 5 nm thick solid Ti film was formed on the surface of the LBO prior to Ni sputtering. The film was necessary for further measurements using a magnetic force microscope (MFM). Further, it was grounded during MFM measurements to exclude the impact of electrostatic interaction between the MFM probe and the sample.

Then Ni was sputtered onto the surface of the substrate in the form of square microparticles with lateral dimensions of $7.5 \,\mu$ m. A metal mesh with appropriate hole sizes was tightly pressed to the surface of the substrate during the Ni deposition for this purpose. The method of electron beam sputtering of a solid-state target made of a corresponding material under ultrahigh vacuum conditions was used for metal sputtering. The substrate temperature was maintained at $+35^{\circ}$ C during sputtering. This made it possible to conditionally cool the sample in the range from +25 to $+35^{\circ}$ C when performing MFM measurements, and heat it in the range from +35 to $+55^{\circ}$ C.

Then, an array of several submicron rectangular particles with the size of $0.9 \times 0.3 \times 0.03 \,\mu$ m was formed using scanning probe lithography from a separate square polycrystalline Ni microparticle with a size of $7.5 \,\mu$ m. All the excess metal was actually scraped off the surface of the substrate with a diamond probe (the so-called "scratching" method) for this purpose. The excess Ni had poor adhesion to the substrate surface and was easily removed by washing the sample with distilled water in ultrasonic baths after separation from the particles. The particles in each of the formed arrays were arranged in such a way that their long side formed a certain angle with the z-axis of crystal LBO substrate (Figure 1). A different number of particles was

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Figure 1. Image of the surface areas of the substrate with formed Ni particles located at angles to the z-axis of the substrate: $(a) - 0^{\circ}$; $(b) - 20^{\circ}$; $(c) - 50^{\circ}$; $(d) - 65^{\circ}$; $(e) - 90^{\circ}$. The scale bar is $1 \mu m$. The height difference for a - 40 nm; b, c - 50 nm; d - 120 nm; e - 60 nm. The arrow shows the direction of the z-axis of the substrate from LiB₃O₅.

obtained in each array since a $7.5\,\mu m$ square microparticle was used to form a separate array. In some cases, the diamond probe detached a particle from the surface of the substrate during the lithography (Figure 1, b, c). The number of particles in each resulting array ranged from 9 to 15. The obtained particles were positioned on a kind of pedestal made of the LBO substrate during the lithography, i.e., because it was difficult to control the depth of penetration of the diamond probe into the substrate and it was necessary to guarantee complete removal of metal between the particles. Therefore, the height of the LBO pedestal for each row of manufactured particles was different, which caused a large height variation observed in AFM images (Figure 1) of the obtained particle arrays. The height of all Ni particles is $0.03\,\mu m$ in this case and is determined by the thickness of the sputtered metal.

The obtained Ni particles were polycrystalline and according to the literature data their coefficient of thermal expansion was $\alpha_p = 1.3 \cdot 10^{-5} \,^{\circ}\text{C}^{-1}$ [14]. An increase of the sample temperature should have resulted in the stretching of the particles along the *x*-axis of the LBO substrate based on the above thermal expansion coefficients. The induced uniaxial deformation with temperature variation (ΔT) should have been $\varepsilon = ((\alpha_x - \alpha_p) - (\alpha_z - \alpha_p)) \cdot T = 7.4 \cdot 10^{-5} \cdot \Delta T$ in this case. According to the literature data [14] the Young's modulus of Ni is equal to E = 210 GPa and the thermally induced uniaxial mechanical stress should have been $\sigma = 15.5 \cdot \Delta T$ MPa. Due to the negative magnetostriction constant of Ni, the stretching of the particles should result



Figure 2. The layout of the particle on the substrate — (a); The arrows show the directions of the axes of the LBO crystal and the external magnetic field (B_{ext}), as well as the direction of magnetization of the particle (M). MFM image: the initial state of Ni particles in the field 0 mT after switching off the magnetizing field -50 mT with the sample temperature of 25° C (b); the initial state of the particles in the field -20 mT with the sample temperature of 35° C (c); in field 0 mT after remagnetization and lowering of the temperature to 25° C — (d). The scale bar is 1μ m. The dependence of the number of particles (N) that changed the direction of magnetization on the external magnetic field and temperature of the structure — (e). The figures (triangle, circle, square) show experimental data, lines (solid, dotted and dots) show fitting results for appropriate temperatures. The dependence of the number of remagnetized particles (dN/dB), is a derivative of the approximation curves (e), from the external magnetic field at different temperatures — (f).

in the creation of an axis of induced magnetic anisotropy in the direction perpendicular to this tension, i.e. along the z-axis. A decrease of the sample temperature should have resulted in the compression of particles along the x-axis of the LBO substrate and, accordingly, in the creation of an axis of induced magnetic anisotropy in the same direction, i.e. perpendicular to the z-axis.

Scanning probe microscope (SPM) Ntegra was used to perform probe lithography and MFM measurements. The probe lithography was performed using diamond probes "D300" (SCDprobes). MFM measurements were performed with low-moment magnetic probes "PPP-LM-MFMR" (Nanosensor). Also, MFM measurements were carried out in a single-pass mode to reduce the impact of the MFM probe on the distribution of magnetization in particles. A permanent magnet integrated into the SPM was used for performing MFM measurements in an external magnetic field, which made it possible to create a magnetic field up to 0.08T in the sample plane. The sample was positioned in such a way during the MFM measurements that the external magnetic field was directed along the *z*-axis of the LBO single crystal. The SPM was also provided with a temperature cell that allowed the sample to be heated from room temperature to 150° C.

The measurement of the magnitude of the particle switching field was performed as follows. A magnetic field significantly larger than the particle anisotropy field (-50 mT) was switched on at a room temperature and the particles were quasi-uniformly magnetized in it. Then the field was switched off and an MFM measurement of the magnetization distribution in the particles was performed. This condition was considered as an initial condition. In this case, both an inhomogeneous distribution of magnetization in particles (Figure 2, *b*) and a quasi-homogeneous distribution of magnetization could be observed (Figure 3, *b*). Next, the sample was heated to the desired temperature, i.e.,

Figure 3. The layout of the particle on the substrate -(a); The arrows show the directions of the axes of the LBO crystal and the external magnetic field (B_{ext}), as well as the direction of magnetization of the particle (M). MFM image: initial state of Ni particles in the field of 0 mT after switching off the magnetizing field of -50 mT at sample temperature of $30^{\circ}\text{C} - (b)$; initial state of particles in the field of -30 mT at sample temperature of 35°C (c); in field 0 mT after remagnetization and lowering of the temperature to $30^{\circ}\text{C} - (d)$. The scale bar is $1 \mu \text{m}$. The dependence of the number of particles (N) that changed the direction of magnetization on the external magnetic field and sample temperature -(e). The figures (triangle, circle, square) show experimental data, lines (solid, dotted and dots) show fitting results for appropriate temperatures. The dependence of the increase of the number of remagnetized particles (dN/dB) is a derivative of the approximation curves (e), from the external magnetic field at different temperatures -(f).

such deformation was created by changing the temperature that affected the magnitude and direction (compression or stretching) of thermally induced uniaxial stresses acting on the particle. After that, the external magnetic field was applied in the opposite direction and gradually increased with a certain increment (1mT). MFM scanning of the studied particle's array was performed at each increment. The external magnetic field increased until the quasihomogeneous magnetization of the particle took place in the field. This value of the external field was taken as the switching field of this particle. The growth of the magnetic field continued until all the particles were magnetized by the field. Then the external magnetic field was switched off and the sample was cooled to room temperature. After that, an MFM image was acquired again (Figure 2, d), which was compared with the initial image of the particle (Figure 2, b). This was done to check the stability of the obtained state. The state was considered stable if the resulting MFM image

was quasi-homogeneous (Figure 3, d) and inverted with respect to the initial one (Figure 3, b).

3. Results and discussions

The measurement of switching fields was started with an array of particles, the long side of which was oriented parallel to the *z*-axis of the LBO substrate (angle 0°). Schematically, the orientation of the particles relative to the substrate and the external magnetic field is shown in Figure 2, *a*. Figure 2, *b* shows the MFM image of particles in the initial state at room temperature (25° C). The state of homogeneous magnetization distribution is not observed in particles. This is attributable to the fact that the particles are formed at a temperature of 35° C. Accordingly, uniaxial compression of the particle takes place at a temperature of 25° C in the direction of the *z*-axis of the LBO substrate and the axis of magnetically elastic anisotropy Averaging experimental value of the switching field (B_{sw}) Ni particles depending on the sample temperature and angle (α) between the long side of the particles and the *z*-axis LBO substrate

	Angle α , (°)	Sample temperature, (°C)						
		25	30	35	40	45	50	55
Switching field B _{sw} , (mT)	0	7	7	10	10	11	11	11
	20	_*	17	21	23	24	29	29
	50	17	15	14	13	14	12	12
	65	_*	20	19	16	13	13	15
	90	_*	47	37	27	20	18	14

Note. *A dash in the table means that no measurements were carried out at this temperature.

is formed in the same direction, which in the absence of an external magnetic field results in the destruction of the homogeneous magnetization of the particle. All particles were uniformly magnetized by an external magnetic field -20 mT (Figure 2, c) and the measurement of the particle switching field was started. The magnetic field was reduced to zero for this purpose and it was increased in the opposite direction in increments of 1 mT. The MFM scanning was performed at each increment and the number of particles was recorded, the direction of magnetization of which was reversed (Figure 2, e). A new cycle of measurement of the particle switching field was carried out at a different temperature after all the particles were remagnetized. The temperature increment was 5°C.

The dependence of the number of switched particles on the magnitude of the applied external field was obtained for each temperature of the sample (Figure 2, e). For clarity, the figure shows the results for only three temperatures: 25, 35 and 45°C. The number of switched particles was adjusted to the total number of particles in the studied array for the convenience of further analysis and to compare the data obtained on different arrays. Since the number of measured particles was small and they had a size spread, these data were approximated by the function $Y = a/(1 + b \cdot \exp(-k \cdot X))$ to average the values obtained. Further, this curve was differentiated and the average value of the particle switching field at a given temperature was determined by the maximum of the derivative (Figure 2, f). The maximum change of the magnitude of the particle switching field is achieved with an increase of the temperature from 30 to 45°C according to the data obtained with such an orientation of the particles relative to the z-axis of the LBO crystal. It increases by 4 mT or 1.57 times in this case.

After measuring the switching field of particles whose long side was parallel to the *z*-axis of the LBO substrate, a similar series of measurements was performed for an array of particles whose long side was located at an angle 65° to this axis. Figure 3, *a* shows the arrangement of such particles on an LBO substrate. These particles in the initial state, unlike the previous ones, had quasi-homogeneous magnetization (Figure 3, b). The direction of magnetization switched under the impact of an external magnetic field which remained after removal of the external magnetic field and cooling of the sample (Figure 3, d). In our opinion, such a stable state of quasi-homogeneous magnetization of particles is attributable to the fact that the axis of induced anisotropy, formed during compression at an angle of 90° to the z-axis of the LBO, is quite close to the direction of the easy axis, the angle $83^{\circ} = 65^{\circ} + 18^{\circ}$, due to the location of the rectangular particle relative to the substrate (angle 18° for the diagonal of an ideal rectangular particle with lateral dimensions of $0.9 \times 0.3 \,\mu$ m and angle of 65° orientation of the long side of the particle to the z-axis of the LBO). The room temperature was about 28°C when measurements were performed on this array of particles, therefore, they were started at a temperature of 30°C for correct comparison with other particles. Figure 3, e, f shows the obtained data on the dependence of the switching field of such particles. The maximum change of the magnitude of the particle switching field is achieved with an increase of the temperature from 30 to 45°C according to the data obtained with such an orientation of the particles relative to the z-axis of the LBO crystal. In this case it decreases by 7 mT or 1.54 times. The data obtained on the value of the particle switching field for different particle orientations and different sample temperatures are presented in the form of a table for convenience.

The data obtained in a similar way for arrays of particles with the long side at angles of 20, 50 and 90° to the *z*-axis of the LBO substrate are presented in the form of graphs in Figure 4. The room temperature for the particle arrays for angles of 20 and 90°, as in the previous case for the array 65° , was higher than 25° C, so measurements were started at a temperature of 30° C. The values of the switching fields for these particles are also provided in the table.

The analysis of the experimental data shows that the switching field increases with the increase of the sample temperature if the angle between the long side of the particles and the z-axis of the LBO substrate is 0° or 20° . This field decreases at angles of 50, 65 and 90° . The change of the switching field for particles of different orientations and at two temperatures of 30 and 45°C is shown in the form a chart in Figure 5. The chart can be conditionally divided into two regions where the opposite behavior of the particle switching field is observed. The value of the switching field of the particle increases with the increase of the temperature at small angles (from 0 to approximately 30°) between the long side of the particle and the z-axis of the LBO substrate. The value of the particle switching field decreases with the increase of the temperature at large angles (from about 50 to 90°).

The observed changes of the particle switching fields are most likely attributable to the following processes. When there is no deformation in the sample and particles are not exposed to any mechanical stresses, the anisotropy

Figure 4. The dependences of the number of particles (*N*) that changed the direction of magnetization on the external magnetic field and sample temperature for particles at an angle of $20^{\circ} - (a)$, $50^{\circ} - (c)$, $90^{\circ} - (e)$. The figures (triangle, circle, square) show experimental data, lines (solid, dashed and dotted) show fitting results. The dependence of the increase of the number of remagnetized particles in the form of a derivative (dN/dB) on the external magnetic field at different temperatures for angles of 20° (b), $50^{\circ} - (d)$, $90^{\circ} - (f)$ by corresponding approximation curves (a, c, e).

of the shape of rectangular particles (K_{sh}) results in the presence of two axes of light magnetization of these particles located diagonally across the rectangle. The particle is compressed in a direction perpendicular to the *z*-axis of the LBO substrate at a sample temperature below its sputtering temperature $(T_{dep} = 35^{\circ}\text{C})$. In this case, the magnetic elastic anisotropy (K_{me}) is induced in the same

Figure 5. The dependences of the mean value of the switching field (B_{sw}) of particles on the orientation of their long side relative to the *z*-axis of the LBO substrate for two temperatures of the sample. The dots are connected by lines for a better perception of the switching field behavior.

direction because of the negative magnetostriction of nickel Accordingly, the direction of effective (Figure 6, a). anisotropy due to these two factors (K_{eff}) at small angles between the long side of the particle and the z-axis of the LBO substrate will be very different from the direction of the external magnetic field (which is applied along the z-axis of the LBO-substrates). Therefore, it will be more difficult to uniformly magnetize a particle in this direction with an external magnetic field, and the quasi-homogeneous magnetization of the particle will collapse when the external field is switched off, which was observed in the experiment (Figure 2, b, d). The particles will begin to stretch in a direction perpendicular to the z-axis of the LBO substrate when the temperature increases above T_{dep} . Accordingly, the magnetic elastic anisotropy (K_{me}) will be induced along the z-axis. This will cause the angle between the directions K_{me} and K_{sh} to become much smaller in particles with small angles (Figure 6, b) and the anisotropy induced in this way will hold the direction of magnetization particles and large values will be needed for their switching by the field, which is observed experimentally (Figure 5).

The particle switching field will exhibit the opposite behavior at large angles between the long side of the particle and the *z*-axis of the LBO substrate. The direction K_{me} will be the same at temperatures below T_{dep} , but the long side of the particle will be at a large angle to the direction of the external magnetic field. Accordingly, the direction K_{sh} will be close to K_{me} and K_{eff} will differ from the direction of the magnetic field by a large angle (Figure 6, *c*). Therefore, the value of the particle switching field will be large. The

Figure 6. The layout of the particle relative to the axes x and z of the LBO crystal. The arrows show the direction of: external magnetic field (\mathbf{B}_{ext} for remagnetization of particles, shape anisotropy (K_{sh}), induced magnetic elastic anisotropy (K_{me}), total (effective) anisotropy (K_{eff}), created mechanical stresses (σ) in particles for small angles of the long side of the particle to the z-axis of the LBO crystal — (a, b), for large angles — (c, d) at temperatures below ($T < T_{dep}$) — (a, c) and above ($T > T_{dep}$) — (b, d) temperatures spraying of the sample.

direction K_{eff} will form an increasingly smaller angle with the direction of the external magnetic field when the sample is heated above T_{dep} particles, which will lead to a decrease of the switching field of the particle (Figure 6, *d*). A further increase of the stretching of the particle can result in the fact that the induced magnetic elasti anisotropy will hold the direction of magnetization of the particle and its switching field will begin to grow again, which is observed experimentally (table) at an angle of 65°. Only a decrease of the switching field of the particle was observed at an angle of 90°. Perhaps the heating temperature of the sample was not enough to achieve the desired value of the tensile stress in the particle.

According to the data obtained (table), the maximum change of the switching field with temperature changes is observed in particles oriented at an angle of 90°. The external magnetic field turns out to be directed along the axis of hard direction of magnetization for such particles at room temperature and therefore the switching field initially has the greatest importance. The uniaxial stretching of the particles along the axis x of the substrate takes place when the sample is heated above 35°C. The magnetic elastic anisotropy is induced in the direction of the z-axis of the substrate due to the negative magnetostriction of Ni, which leads to a rotation of the easy axis of the particle closer to this direction (Figure 6, d) and a strong decrease of its switching field in this direction. The maximum decrease of the switching field in the case of an angle of 90° by 3.36 times is observed with an increase of temperature

from 30 to 55° C and equals to 33 mT. Despite the largest effect on the reduction of the switching field among the studied particle orientations, the use of such an arrangement is complicated, since switching off the external magnetic field after their remagnetization and cooling of the sample to room temperature does not lead to an unambiguous rotation of the magnetization by 180° relative to the initial state. This process is probabilistic in nature, since both orientations of magnetization are equally probable.

4. Conclusion

According to the data obtained, it is possible to reduce or increase the magnitude of the switching field in the studied rectangular Ni particles by choosing the direction of uniaxial mechanical stress acting on the particle. The thermally induced stretching of particles at angles of 60-90° to their long side and the application of an external magnetic field at an angle of $0-30^{\circ}$ to their long side results in an increase of the particle switching field. Thermally induced stretching of particles at angles of $0-40^{\circ}$ to their long side and the application of an external magnetic field at an angle of $50-90^{\circ}$ to their long side leads to a decrease of the switching field of particles. Thermally induced stresses have little effect on the particle switching field in the remaining range of the angles. The strongest decrease of the particle switching field is observed if the long side of the particle is parallel to the direction of thermally induced stretching, and the direction of the external field is perpendicular to it. In this case, the switching field decreases by 3.36 times (from 47 to 14 mt) when the substrate temperature changes by $25^{\circ}C$ (from 30 to $55^{\circ}C$).

A rotation of the direction of magnetization from a state along the external magnetic field to a state along the axis of light magnetization of particles attributable to their shape anisotropy was observed in the particles formed at angles 0, 20, 50 and 65° after switching off the magnetic field and removing induced deformations (i.e. at a temperature of 35° C). The obtained magnetization differed from the initial one by $\approx 180^{\circ}$ because of this.

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Conflict of interest

The authors declare that they have no conflict of interest.

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