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BaM hexaferrite ($BaFe_{12}O_{19}$) thin films on Al_2O_3(01-12) substrates: crystal structure and magnetic properties

© B.B. Krichevtsov[1], A.M. Korovin[1], S.M. Suturin[1], A.A. Levin[1], N.S. Sokolov[1], V.V. Fedorov[2], A.V. Telegin[3], D.A. Shishkin[3]

[1] Ioffe Institute,
St. Petersburg, Russia
[2] Alferov Federal State Budgetary Institution of Higher Education and Science Saint Petersburg National Research Academic University of the Russian Academy of Sciences,
St. Petersburg, Russia
[3] M.N. Mikheev Institute of Metal Physics, Ural Branch, Russian Academy of Sciences,
Yekaterinburg, Russia

e-mail: boris@mail.ioffe.ru

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Epitaxial films of BaM hexaferrite ($BaFe_{12}O_{19}$) with a thickness of 50 nm were grown on R-cut sapphire α-Al_2O_3 (01−12) substrates by laser molecular beam epitaxy (LMBE). Their crystal structure and magnetic properties were studied before and after post-growth annealing. It was found that in the annealed films obtained by the LMBE method, the easy axis of magnetic anisotropy is deviated from the normal to the surface, which makes it possible to switch the magnetization with both normal and tangential magnetic fields, and leads to the dependence of the hysteresis loop shape on the field orientation in the film plane.

Keywords: hexaferrites, thin films, magnetization processes, laser molecular beam epitaxy.

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Limitations in the speed and volume of transmitted information in modern computer chips, whose action is based on using electric currents, are due to the release of Joule heat. As shown in a number of studies, this problem can be solved by using spin waves (SW) to transmit and process information. For the excitation, control and reception of SWs thin film magnetic materials with low attenuation of SWs, the ability to control their direction and operate at frequencies $\sim 50-100$ GHz are required. Promising materials for these purposes are hexaferrites, in particular, hexaferrite BaM ($BaFe_{12}O_{19}$), which has high values of uniaxial anisotropy field $H_a \sim 18$ kOe and saturation magnetization $4\pi M_s \sim 4.6$ kG, allowing to implement devices at such frequencies. The presence of strong uniaxial magnetic anisotropy makes it possible to obtain thin films of BaM hexaferrite in which, in the absence of a magnetic field **H**, a monodomain state with the orientation of the magnetization normal to the plane of the structure [3–6].

It is important to note that structures in which, in the absence of a field **H**, a state with magnetization **M** lying in the plane of the film or directed at an angle to it [7,8] are also interesting for practical applications. In the latter case, magnetization can be switched over by a magnetic field oriented either in the plane of the structure ($\mathbf{H}_{\text{in-plane}}$) or normal to this plane ($\mathbf{H}_{\text{out-of-plane}}$). Such a state can be implemented by an appropriate choice of the substrate [8]. In this study, BaM hexaferrite films were grown by laser molecular beam epitaxy (LMBE) on R-cut Al_2O_3 substrates and the process of magnetization switching at different orientations of the magnetic field **H** was investigated.

BaM hexaferrite films were grown on Al_2O_3(01−12), substrates, in which the direction of the C_3 axis is oriented at the $\sim 62°$ angle to the plane. The films were grown at $T_{gr} = 700°$C temperature and $p = 0.06$ mbar oxygen pressure. After growth, the structures were removed from the growth chamber and annealed in a muffle furnace at $T_{ann} = 1000°$C for one hour. The film thickness was 50 nm. The growth rate was calibrated using quartz scales. The crystal structure of the films was monitored by mapping of reflection high-energy electron diffraction (RHEED). The surface morphology of films and substrates was investigated using an INTEGRA atomic force microscope (NT-MDT LLC, Zelenograd, Russia).

X-ray diffraction (XRD) measurements were performed in the $\theta-2\theta$ scanning mode on a D2 Phaser powder X-ray diffractometer (Bruker AXS, Karlsruhe, Germany) equipped with a LYNXEYE linear semiconductor position-sensitive detector (Bruker AXS) in the vertical $\theta-\theta$ Bragg−Brentano geometry. Cu-K_α-radiation (wavelength $\lambda = 1.5418$ Å) of an X-ray tube with a copper anode filtered by a nickel foil filter was used.

Moreover, XRD studies were carried out using reciprocal space mapping. To do this, a Super Nova diffractometer (Agilent Technologies, Inc., USA) with a two-dimensional (2D) detector (Atlas S2 CCD) and a copper anode X-ray emitter ($\lambda = 1.5418$ Å) was used. A 3D diffraction pattern

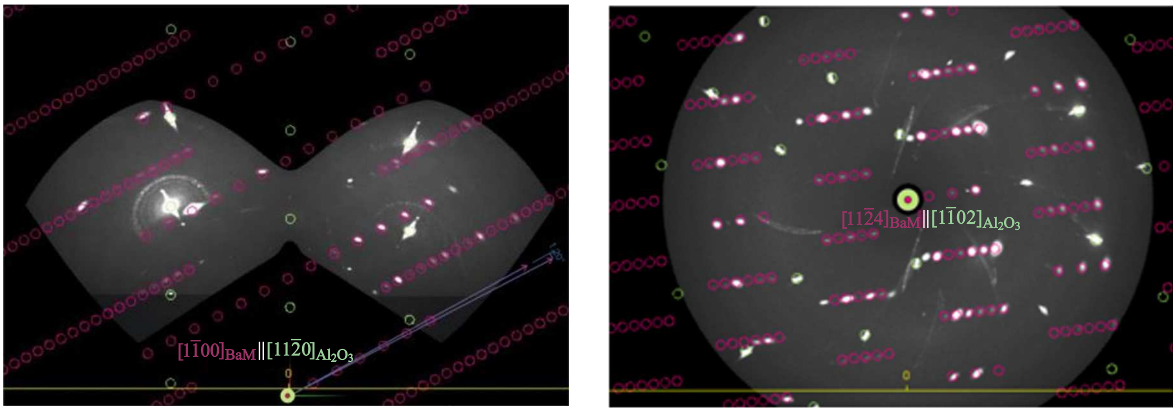

Figure 1. Two perpendicular reciprocal space cross sections captured by XRD in a BaM hexaferrite sample grown on $Al_2O_3(1-102)$ and annealed. Red circles — model positions of the BaM hexaferrite lattice reflexes. Green circles correspond to the substrate Al_2O_3. The red and green numbers on the axes refer to the film and substrate, respectively.

mapping method was used to analyse the RHEED and XRD patterns obtained for different θ diffraction patterns.

The magnetic hysteresis loops of the obtained structures, were measured using vibrational magnetometry (VSM) on a Lake Shore 7400 Cryotronics (Lake Shore Cryotronics, Inc., USA). The magnetic field **H** was applied out-of-plane, or at various angles (azimuths) in-plane. In both cases, the magnetic moment component ($\mathbf{M}_{\text{out-of-plane}}$ or $\mathbf{M}_{\text{in-plane}}$), parallel to the magnetic field ($\mathbf{H}_{\text{out-of-plane}}$ or $\mathbf{H}_{\text{in-plane}}$ respectively) was measured.

In addition, laser polarimetry setup ($\lambda = 405$ nm) of polar magneto-optical Kerr effect (PMOKE) was used to measure the loops. Magnetization curves were studied at magnetic field orientations **H** out-of-plane of the structure, and in-plane of the structure. In the latter case, the dependences of the magnetization component out-of-plane on the magnetic field oriented in-plane were measured: $\mathbf{M}_{\text{out-of-plane}}(\mathbf{H}_{\text{in-plane}})$.

Image analysis of the surface of the annealed structure with a thickness of 50 nm showed that the film consists of nanocrystallites with lateral dimensions of $\sim (200-300)$ nm. The crystallites are densely packed, but there are rather deep voids between some of them, reaching almost to the substrate. The average roughness of such a surface is $RMS = 15.8$ nm over an area of $5 \times 5\,\mu$m.

As shown by studies of films grown on $Al_2O_3(0001)$, C-cut sapphire substrates, the crystal structure of BaM hexaferrite is formed only after annealing in air [5,6]. The same situation is observed during the growth of $BaFe_{12}O_{19}$ on R-cut sapphire substrates. The results of crystal structure investigation by RHEED and XRD indicate that in the structures $BaFe_{12}O_{19}/Al_2O_3(01-12)$, as in $BaFe_{12}O_{19}/Al_2O_3(0001)$ [5,6], the crystal lattice of BaM hexaferrite (PDF-2 01-075-9113) is only implemented after annealing in air at $T = 1000^\circ$C (Fig. 1).

The simulation of XRD diffraction patterns showed that the film grown on the R-cut sapphire substrate $Al_2O_3(01-12)$ yields a monodomain structure of BaM hexaferrite of high crystalline quality after annealing, which lacks polycrystallinity and texture. All reflections are attributed to either the substrate or the BaM film and the structure lacks other phases and texture (reversals around the normal). Epitaxial ratios film-structure: direction $[1-102]_{Al_2O_3}$ is parallel $[11-24]$ in the substrate plane, direction $[11-20]_{Al_2O_3}$ is almost parallel to the direction $[22-4-1]_{BaM}$ with $\sim 1.2^\circ$ turning around the axis $[1-100]_{BaM}$. The hexagonal axis of the film is deflected from the normal to the surface by the angle $\varphi \sim 62^\circ$.

As in the films grown on $Al_2O_3(0001)$, substrates, the magnetic properties in $BaFe_{12}O_{19}/Al_2O_3(01-12)$ structures only appear after annealing in air. The hysteresis loops in the $\mathbf{H}_{\text{out-of-plane}}$ field measured by PMOKE and VSM are shown in Fig. 2. The PMOKE loops at $\mathbf{H}_{\text{out-of-plane}} = -5-+20$ kOe and $+5--20$ kOe are formed by smooth, reversible $M_{\text{out-of-plane}}(H_{\text{out-of-plane}})$ dependences associated with magnetization rotation to the magnetic field direction, and irreversible magnetization jumps at $H \sim \pm 5$ kOe caused by the formation and movement of domain walls. Similar jumps are also observed in the loops obtained with VSM (Fig. 2, *b*). Note that after subtracting the linear in H contribution, no magnetization rotation is evident in these loops. This is due to the strong contribution of the substrate susceptibility to the loops measured with VSM, which is not evident in the PMOKE loops because light at 405 nm is strongly absorbed in the film. The value of the residual magnetization in the PMOKE loops shows that in the absence of a magnetic field, the films realise a monodomain state with a magnetization orientation **M** approximately along the C_3 axis of the substrate. This confirms the XRD results in that the hexagonal axis and consequently the light magnetization axis in the films are oriented at an angle $\sim 62^\circ$ to the substrate plane.

The magnetic hysteresis loops in the $\mathbf{H}_{\text{in-plane}}$ magnetic field measured by PMOKE and VSM show a strong dependence of the loop shape H_c coercive field and M_{rem} residual magnetization on the magnetic field azimuth θ in the plane of the structure (Fig. 3). Note that in

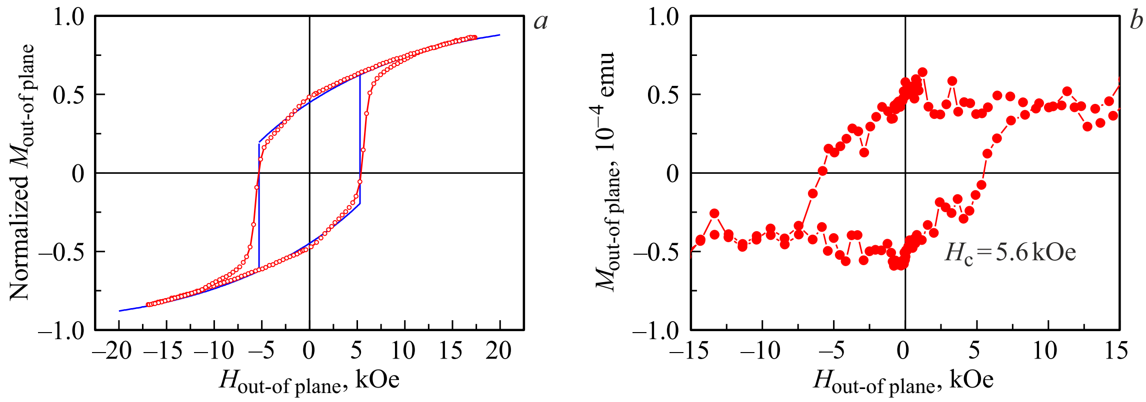

Figure 2. (a) Hysteresis loop measured with PMOKE in the $\mathbf{H}_{\text{out-of-plane}}$ field (red circles). Blue solid curve — results of the loop calculation using the Stoner–Wohlfarth model for values of $4\pi M_s = 4.5$ kG, $H_a = 18$ kOe, $\varphi = 62°$. (b) Hysteresis loop measured with VSM after subtracting the linear in H part manifested in strong fields.

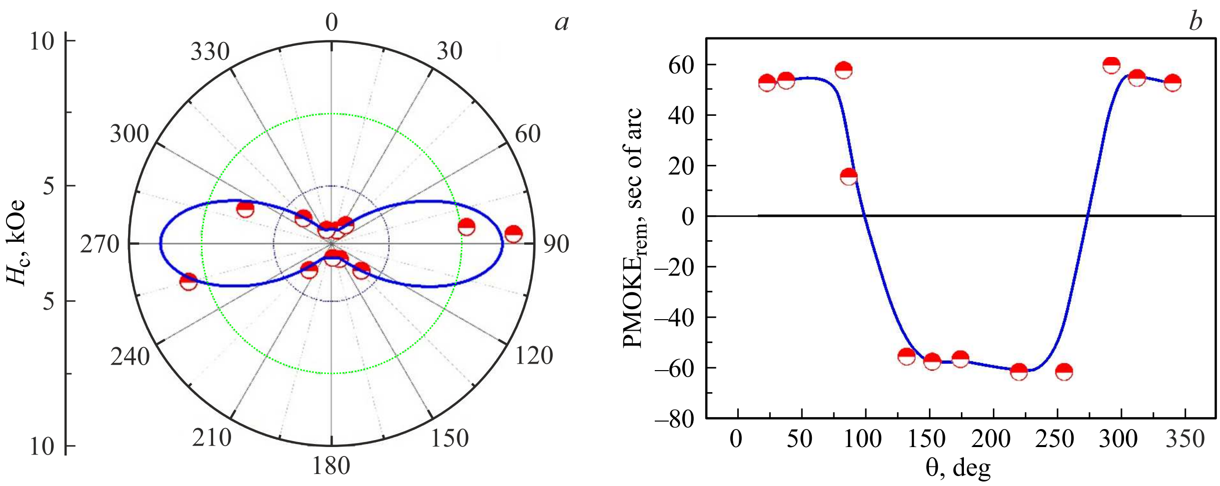

Figure 3. (a) Dependence of the coercive field Hc on the magnetic field azimuth in the θ plane obtained from PMOKE loops. (b) Angular dependence of the residual magnitude of $\text{PMOKE}_{\text{rem}}$. Blue solid lines are shown for visual convenience.

the case of PMOKE the measured loops are due to $M_{\text{out-of-plane}}(H_{\text{in-plane}})$ dependencies while in the case of VSM — $M_{\text{in-plane}}(H_{\text{in-plane}})$. The anisotropy of the loops is due to the fact that as the azimuth of the magnetic field θ changes, the projection of the easy magnetic axis (EMA) to the direction of the magnetic field $\mathbf{H}_{\text{in-plane}}$ also changes. As a result, magnetization jumps should be observed when this projection is different from zero. If the magnetic field is perpendicular to the EMA, only magnetization rotations should be observed, which is what appears in Fig. 3. At $\theta = 0°$ and $180°$ the EMA projection is parallel to the magnetic field and a narrow loop with $H_c = 3.5$ kOe and a large residual PMOKE value is observed. At $\theta = 90°$ and $270°$ the residual value of PMOKE changes practically by a jump with a change of sign, which reflects the change of the EMA projection to the magnetic field direction (Fig. 3, b).

Thus, thin films of BaM hexaferrite grown by laser molecular beam epitaxy on $Al_2O_3(01-12)$ substrates after annealing in air show the crystal structure of BaM hexaferrite with the direction of the sixth-order axis at the angle $\sim 62°$ to the plane and uniaxial magnetic anisotropy with the direction of the EMA along this axis. In the absence of a magnetic field, a monodomain state with the magnetization direction near the EMA is implemented. Magnetic hysteresis loops are observed in a field orientated both in-plane and out-of-plane. The anisotropy of the magnetic hysteresis loops was found when the film is remagnetised by a tangential magnetic field. It is shown that the magnetization direction can be changed within a wide range by a magnetic field oriented both out-of-plane and in-plane of the structure, which expands the possibilities of applying such films in VHF devices.

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Conflict of interest

The authors declare that they have no conflict of interest.

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