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Magnetron Deposition of Barium Hexaferrite Films from a Target of Stoichiometric Composition

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Abstract—A technique is proposed for obtaining films of barium hexaferrite on sapphire with (0001) orientation by high-frequency magnetron sputtering of a stoichiometric $\text{BaFe}_{12}\text{O}_{19}$ target followed by annealing. The X-ray diffraction analysis and Raman spectrometry carried out showed the possibility of crystallization of oriented films without using correction of the target composition.

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Compounds of iron oxide with oxides of other metals, called ferrites, form various groups that are combined according to a structural feature. For example, garnet ferrites, orthoferrites, spinel ferrites, and hexaferrites. The last two groups, in particular, can be represented by barium ferrites. The first mention of the study of barium ferrite dates back to 1931 [1]. The discovery of hexaferrites in the 1950s gave impetus to the intensification of research into these compounds. The most famous of them is barium hexaferrite. Like all hexagonal ferrites, it is a ferrimagnet.

Today, the increased commercial and technological interest in this material is due to its possible use in the elements of devices operating at gigahertz frequencies. These are areas such as mobile and wireless communications, data recording and storage, and electromagnetic wave absorbers. The development of nanotechnology dictates the need to develop reproducible methods for obtaining the necessary materials in the form of thin-film elements. Various methods for successfully obtaining barium hexaferrite films have been reported: pulsed laser deposition [2], ion beam sputtering [3], molecular beam epitaxy [4], liquid phase epitaxy [5], etc. At present, for this purpose, magnetron deposition methods on direct and alternating current are used more often than others [6, 7]. In this case, the first option requires the use of special low-resistance targets. The use of ceramic targets made of stoichiometric ceramics in reactive processes is difficult due to the possible bombardment of the growing film with negative oxygen ions, and, as a result, back-sputtering of barium atoms. This leads to a deviation of the resulting film composition from the stoichiometric

one. To compensate for losses, the authors of [6] placed elements enriched with barium on the target.

In this study, we used sputtering of a stoichiometric target in an argon atmosphere onto a cold substrate. The minimum oxygen content in the vacuum chamber during target sputtering makes it possible to avoid bombardment of the growing layer with negative oxygen ions, and the low temperature of the substrate prevents the selective re-evaporation of deposited atoms, which has a positive effect on the transfer of the target composition to the substrate. The pressure in the chamber was 3×10^{-3} Torr, target diameter 50 mm, and distance to the substrate of 50 mm. At a discharge power of 100 W, the deposition rate was 0.25 $\mu\text{m}/\text{h}$.

The resulting films were studied by X-ray diffraction and Raman spectroscopy. X-ray diffraction analysis was carried out on a DRON-3 automated diffractometer with a quartz monochromator. The wavelength of the X-ray tube was 0.15405 nm. The survey was carried out in Bragg–Brentano geometry. The Raman spectra were recorded using an EnSpectr M532 Raman microscope with a laser wavelength of 532 nm. Immediately after deposition, no pronounced peaks were found in the X-ray diffraction pattern, except for the peak from the substrate.

The Raman spectrum is shown in Fig. 1. Very broad peaks are observed, which coincide in position with the spectra for $\text{BaFe}_{12}\text{O}_{19}$ given in [8]. All this indicates that, under the specified conditions of deposition, firstly, the transfer of the sprayed material occurs without composition distortion, and secondly, an amorphous phase is formed. To recrystallize the deposited layer, a series of annealings was carried out at different temperatures. Up to 700°C, no changes in

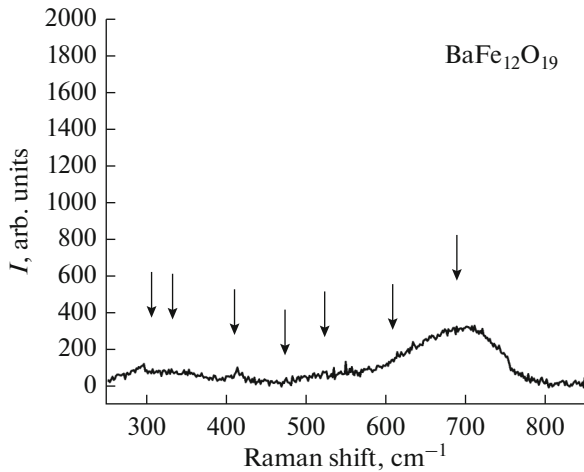


Fig. 1. Raman spectrum from an amorphous film of barium hexaferrite.

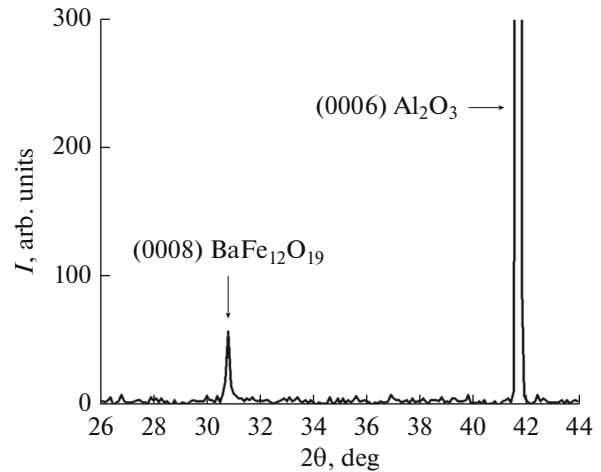


Fig. 2. X-ray spectrum from a film of barium hexaferrite after annealing at 800°C.

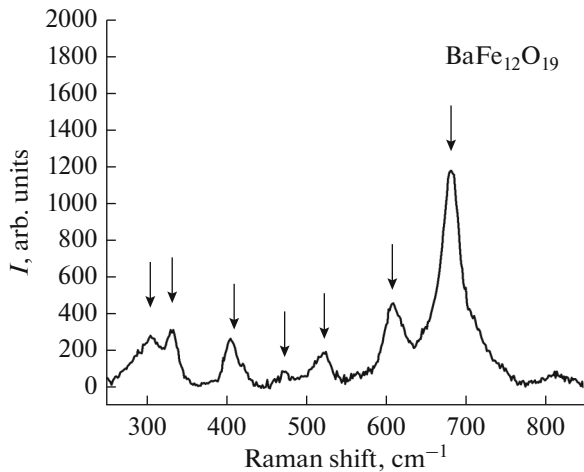


Fig. 3. Raman spectrum from a film of barium hexaferrite after annealing at 800°C.

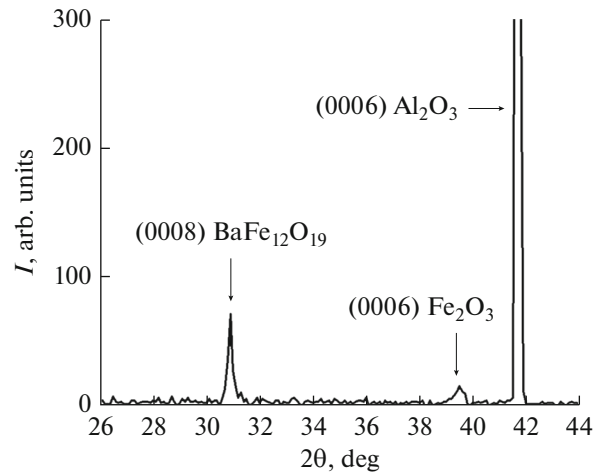


Fig. 4. X-ray spectrum from a film of barium hexaferrite after annealing at 900°C.

the crystal structure were observed. After annealing at a temperature of 800°C, the diffraction pattern (Fig. 2) shows a peak corresponding to the reflection (0008) of $\text{BaFe}_{12}\text{O}_{19}$. From the width of this peak, using the Scherrer method, the size of the coherent scattering regions was determined, which was about 17 nm. The Raman spectrum (Fig. 3) shows the presence of narrow lines, which confirms the successful recrystallization of barium hexaferrite. No foreign phases were found. However, after an increase in the annealing temperature to 900°C, a peak was observed in the X-ray diffraction pattern of the obtained film, which can be interpreted as a reflection from plane (0001) Fe_2O_3 . This indicates that in this case the decomposition of hexaferrite occurs with the release of hematite

(Fe_2O_3) in a separate phase, so we can say that the optimal annealing temperature is 800°C.

Thus, the optimal conditions for obtaining films of barium hexaferrite by magnetron sputtering of a target of stoichiometric composition in pure argon without heating the substrate, followed by annealing in the atmosphere at 800°C. It is shown that in this case there is no need to correct the target composition. And subsequent annealing allows the recrystallization of the obtained amorphous film.

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CONFLICT OF INTEREST

The author declares that he has no conflicts of interest.

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REFERENCES

1. J. Guillissen and Pierre J. Van Rysselberghe, *J. Electrochem. Soc.* **59** (1), 95 (1931).
2. Z. Cai, Z. Chen, T. L. Goodrich, et al., *J. Cryst. Growth.* **307**, 321 (2007).
3. V. G. Kostishin, A. Yu. Mironovich, R. I. Shakirzyanov, et al., *J. Magn. Magn. Mater.* **527**, 167786 (2021).
4. P. R. Ohodnicki, K. Y. Goh, M. E. McHenry, et al., *J. Appl. Phys.* **103** (7), E514 (2008).
5. S. D. Yoon and C. Vittoria, *J. Appl. Phys.* **96**, 2131 (2004).
6. Zh. Xiaozhi, Y. Zhenxing, M. Siqin, and Y. Lixin, *J. Appl. Phys.* **116**, 243909 (2014).
7. A. R. Abuzir and S. A. Salman, *Results Phys.* **8**, 587 (2018).
8. Kreisel J., Pignard S., Vincent H., Sénateur J.P., *J. Appl. Phys. Lett.* **73** (9), 1194 (1998).