Dielectric and magnetic properties of Sr$_3$Co$_2$Fe$_{24}$O$_{41}$ thin hexaferite samples in the microwave range

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Abstract. We present studies on the changes of the dielectric properties of 0.7 – 1.2-mm thick-layers of Sr$_3$Co$_2$Fe$_{24}$O$_{41}$ Z-type hexaferite in the frequency range 5 – 40 GHz; to the best of our knowledge, for the first time we used a combination of two methods – a resonance method, which yields information on the dielectric and magnetic anisotropies of the samples, and a broadband transmission-line method – covered coplanar and microstrip lines. We measured an increase of the dielectric constant and dielectric loss tangent above 20 GHz and a measurable anisotropy of the permittivity and permeability. The changes in the two parameters were also investigated (a magneto-electric effect was observed) in an external dc magnetic field in directions parallel and perpendicular to the sample at room temperature. The investigations presented are complex and intricate and should be considered as a first step in the characterization of thick Sr$_3$Co$_2$Fe$_{24}$O$_{41}$ Z-type hexaferite slabs above 3 GHz up to the mm-wave range.

1. Introduction

The modern microwave ferrites, e.g. spinel, garnet, and hexaferite systems in the form of thin or thick films, powders, metamaterials, and, lately, different multiferroic materials, are key components in the systems that send, receive, and manipulate electromagnetic signals across a very wide frequency range, from VHF up to millimeter-wave bands [1]. The magneto-electric effect (ME) in the multiferroic materials, i.e. permittivity variation in an external dc magnetic field, and vice versa – permeability variation in an external dc electric field, is a remarkable and promising property for applications in various tunable microwave devices [2].

Thin and thick layers of Ba-Sr hexaferites have exhibited such properties [3-5]; one such example is the Ba$_{1.5}$Sr$_{1.5}$Co$_2$Fe$_{24}$O$_{41}$ hexaferite. The substitution of Sr$^{2+}$ for Ba$^{2+}$ was reported as reducing the sintering temperature from 1250 °C to 1210 °C, decreasing the oxygen partial pressure in synthesizing Co$_2$ Z-type ferrite Sr$_3$Co$_2$Fe$_{24}$O$_{41}$ and considerably cutting the costs [3]. Moreover, this substitution also improved the frequency characteristics of the permeability. The dielectric constant of the Z-type hexaferite Sr$_3$Co$_2$Fe$_{24}$O$_{41}$ varies widely for frequencies below 10 MHz, and for temperatures below 100 K [3]. Changes of ~18% in the remanent magnetization have been observed in an electric field of 10 kV/cm. Similarly, a change of ~16% has been measured in the permittivity at 1 GHz in a magnetic

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field of only 320 Oe [4]. Resonance behavior of the dielectric properties has been observed within the 1 – 3 GHz range [4].

In this paper, we describe initial studies on the dielectric and magnetic properties of samples of the Z-type hexaferrite Sr₃Co₂Fe₃O₁₄ in the frequency range 5 – 40 GHz; to our best knowledge, for the first time we used a combination of two methods — a resonance method [6], which allows one to determine the samples’ dielectric and magnetic anisotropy, and a broadband transmission-line method [7] — covered coplanar and microstrip lines.

2. Methods and samples

2.1. Samples

The Sr₃Co₂Fe₃O₁₄ powders were synthesized by sol-gel auto-combustion. The metal nitrates Sr(NO₃)₂, Co(NO₃)₂.6H₂O and Fe(NO₃)₃.9H₂O were used as starting materials and sugar was used as a fuel. The solution was slowly evaporated to form a gel. During the dehydration process, the gel turned into a fluffy mass and burned in a self-propagating combustion manner. During the auto-combustion process, the burning gel volume expanded rapidly and NOₓ gas resulting from the decomposition of nitrate ions was released. The material thus produced was ground and annealed at 600 °C. The precursor material obtained was pressed into disk-shaped pellets, synthesized at 1200 °C for seven hours and then quenched rapidly to room temperature to prevent the formation of other hexaferrite phases. In order to perform reliable measurements in the microwave range, two disk samples under test (SUTs) were prepared: S1 (diameter dₛ = 11.96; thickness tₛ = 1.2 mm) and S2 (dₛ = 9.66; tₛ = 0.67 mm).

2.2. Resonance method for extraction of dielectric and magnetic parameters

The resonance measurements on the hexaferrite SUTs were performed by the two-resonator method described in [6, 8]. Figure 1 schematically illustrates the pair of measurement cylinder resonators used; the disk samples are with diameters smaller than the resonators’ diameter and height so that they can be placed inside the measurement cavities in a horizontal (h) or a vertical (v) position. The measurement resonators were designed to support either symmetrical TEₘₙₙ modes with electric field orientated perpendicular to the axis (in resonator R1), or symmetrical TMₘₙₙ modes with electric field orientated along the axis (in resonator R2). Therefore, the SUT influences the resonance frequency and quality factor (Q) either in a parallel or in a perpendicular direction to the existing E field, so that the applied measurement method can provide the actual anisotropy of the dielectric and magnetic constants and loss tangents, i.e., values parallel (εₓ/tanδₓ and μₓ/tanδₓ) and perpendicular (εᵧ/tanδᵧ and μᵧ/tanδᵧ) to the sample surface.

The SUTs parameters were extracted based on full-wave simulations (ANSYS HFSS®) after measurement of resonance characteristics in both resonators R1/R2 with SUTs that support symmetrical TEₘₙₙ or TMₘₙₙ modes (e.g. usually the first three modes with index m = 1, 2, 3). The TE

![Figure 1](image-url)  
**Figure 1.** Measurement structures and samples: a) Resonator R1 with TEₘₙₙ (m=1,2,3, n=1,2); b) Resonator R2 with TMₘₙₙ modes (m=1,2,3); c) SUT: S1 (dₛ = 11.96; tₛ = 1.2 mm); S2 (dₛ = 9.66; tₛ = 0.67 mm); d) three planar waveguides covered by SUT: coplanar waveguide CPW, microstrip line MSL and thin tMSL. Legend: 1h,v – sample placed horizontally or vertically; 2 – resonator; 3 – foam support; 4 – substrate; red arrows – electric field E directions of exited TE or TM modes in resonators or dominant modes in planar waveguides.
modes with index \( n = 1, 3 \) (with \( E \) field maximum in the SUT) ensure determination of the parallel dielectric parameters \( \varepsilon_e, \tan \delta_e \) when the sample is placed horizontally, or the perpendicular dielectric parameters \( \varepsilon_e, \tan \delta_e \), when the sample is placed vertically—figures 2 a, b. The TE modes with \( n = 2, 4, 6 \) (with \( E \) field minimum in SUT) ensure determination of the purely perpendicular magnetic parameters \( \mu_m, \tan \delta_m \) for a horizontal sample or \( \mu_m, \tan \delta_m \) for a vertical sample. Finally, in a similar way, the TM modes with index \( m = 1, 2, 3 \) allow determination of the perpendicular dielectric parameters \( \varepsilon_e, \tan \delta_e \) for a horizontal sample, and the parallel dielectric parameters \( \varepsilon_e, \tan \delta_e \) for a vertical sample. This measurement procedure ensures possibilities for characterizing hexaferrite SUTs in a wide frequency range 6 – 38 GHz.

\[ \text{Figure 2. Simulated electric } E \text{ and magnetic } H \text{ field distribution in cylinder resonator R1 for TE}_{011} \text{ mode (a, b) and in a 50-ohm CPW (c) and MSL (d) planar structures with sample placed horizontally or vertically.} \]

2.3. Broadband method for parameters verifications by covering a 50-ohm microstrip line

The method considers a measurement structure where the hexaferrite SUT covers 50-ohm planar transmission lines: coplanar waveguide (CPW) or microstrip line (MSL or tMSL) with different conductor widths—figure 1d. We have already applied similar structures for characterization of metamaterials [9]. The dominant modes in both structures, CPW and MSL, have electric \( E \) and magnetic \( H \) field located high above the conductor layout (illustratively presented in figure 2c, d) and penetrating into the hexaferrite sample. Thus, the SUT introduces insertion losses and affects the phase in the entire transmission lines; their frequency dependencies are representative measures of the SUT dielectric and magnetic parameters, when a reliable extraction procedure has been performed (details have been presented in [9]). In fact, the \( E \) field in the CPW are orientated predominantly in parallel to the waveguide substrate; therefore, the CPW \( S \) parameters are influenced mainly by the pairs \( \varepsilon_e, \tan \delta_e \) and \( \mu_m, \tan \delta_m \). MSL has a mixed distribution of the \( E \) field (neither purely parallel, nor purely perpendicular); therefore, the covered MSL allows extraction of a mixed (equivalent) value between the parallel and perpendicular dielectric and magnetic constants. In our case, we simply introduce the parameters obtained by resonance measurements in the 3-D models of the measurement structures and compare the simulated dependencies with the measured ones for all samples considered. However, there exists a measurement problem; usually an unavoidable air gap with an effective thickness \( g_a \) appears between the sample bottom surface and the top CPW and MSL conductors due to various reasons: small curvatures, roughness, surface irregularities, etc. This effective gap can be evaluated by using isotropic samples: \( g_a \sim 2 \pm 0.25 \) \( \mu \text{m} \) for MSL; \( g_a \sim 3.5 \pm 0.5 \) \( \mu \text{m} \) for tMSL; and \( g_a \sim 15 \pm 2.5 \) \( \mu \text{m} \) for CPW.

3. Results and discussion

The resonance measurements of samples S1 and S2 were performed for different modes in a set of cylinder resonator with diameters 30 mm, 18 mm, 15 mm and 10 mm. Figure 3 presents resonance curves of four selected TE modes in R1 and TM modes in R2. The relatively big dielectric constant and loss tangent of the hexaferrite samples studied make it difficult to observe and identify some of the modes (e.g. symmetrical TE modes in R1) due to the low \( Q \) factors (especially for the bigger sample S1). However, we succeeded in obtaining satisfactory results following the procedure described in [8]. The extracted pairs of values \( \varepsilon_e/\tan \delta_e; \varepsilon_e/\tan \delta_e; \mu_m/\tan \delta_m \) and \( \mu_m/\tan \delta_m \) are...
presented in table 1 for selected modes in the frequency range 6 – 24 GHz. The procedure for evaluating the dielectric and magnetic parameters is as follows: First, making use of the corresponding TE and TM modes for samples in a horizontal and a vertical position, we determine coarsely the purely dielectric parameters; then, for modes TE\(_{012, 04, 016}\) with an \(E\) minimum in the SUT, we determine the corresponding purely magnetic parameters with a sufficient accuracy. Finally, using the already determined magnetic parameters in a given frequency range, we recalculate the dielectric parameters obtained using the TE and TM modes with \(E\) field maximum in the sample body. These final values are presented in table 1 in black (the intermediate results are in grey). The last row in table 1 gives the averaged parameters of the hexaferrite samples in the interval 7 – 25 GHz; these values were used in the simulations of covered CPW, MSL and tMSL structures.

Figure 4a presents the measured and simulated frequency dependencies of the additional losses and the additional phase delay (due to the near presence of SUT over the transmission-line conductors) of covered planar waveguides for sample S2 only; the effective air gaps are accounted for in the simulated dependencies. The curves are visibly nonlinear. This fact means that both the dielectric and magnetic parameters of the hexaferrite considered do not remain constant as the frequency is raised, which is the main

![Figure 3](image-url)  
**Figure 3.** Measured resonance curves of selected modes in resonators R1 (TE\(_{011}\); TE\(_{012}\); TE\(_{014}\)) and R2 (TM\(_{010}\)).

**Table 1.** Extracted values of the dielectric and magnetic parameters of hexaferrite sample S2 by resonance measurements of different TE and TM modes for horizontally and vertically placed samples.

| Resonator and mode | Sample position | Resonance frequency / \(Q\) factor | Dielectric parameters \(\varepsilon / \tan\delta_e\) | Magnetic parameters \(\mu / \tan\delta_m\) |
|--------------------|----------------|----------------------------------|---------------------------------|-----------------------------------|
| R1 (\(D = 30\) mm) | horizontal     | 12.4008/31.86                    | Par.: 15.02/0.076              | Perp.: 0.96/0.32                  |
| R1 (\(D = 30\) mm) | vertical       | 13.0374/85.08                    | Perp.: 9.03/0.25               | Perp.: 0.70/0.32                  |
| R2 (\(D = 30\) mm) | h              | 7.5620/494.2                     | Perp.: 9.92/0.445             | Perp.: 0.70/0.30                  |
| R2 (\(D = 30\) mm) | v              | 6.9632/194.5                     | Par.: 14.76/0.026             | Perp.: 0.96/0.32                  |
| R1 (\(D = 30\) mm) | h              | 15.6616/550.6                    | Perp.: 15.18/0.155            | Perp.: 0.92/0.70                  |
| R1 (\(D = 30\) mm) | v              | 15.6681/387.2                    | Perp.: 7.95/0.47              | Perp.: 0.70/0.50                  |
| R1 (\(D = 30\) mm) | h              | 23.3829/963.1                    | Par.: 14.20/0.07              | Perp.: 1.03/0.12                  |
| R1 (\(D = 30\) mm) | v              | 23.3907/511.0                    | Parp.: 8.80/0.10              | Perp.: 0.73/0.40                  |
| R1 (\(D = \)18 mm)| h              | 21.2149/20.75                    | Par.: 14.2/0.145              | Perp.: 0.95/0.70                  |
| R1 (\(D = \)18 mm)| v              | 21.5624/70.18                    | Perp.: 9.25/0.20              | Perp.: 0.69/0.88                  |
| R2 (\(D = \)18 mm)| h              | 12.3280/132.8                    | Perp.: 8.5/0.80               | Perp.: 0.77/0.22                  |
| R2 (\(D = \)18 mm)| v              | 10.2358/61.13                    | Par.: 14.0/0.055              | Perp.: 0.98/0.25                  |
| Averaged           | (7 – 2.5 GHz)  | Par.: 14.7/0.08                  | Perp.: 0.75/0.20              | Perp.: 0.98/0.10                  |
reason why the simulated and measured dependencies do not fully coincide. The observed curvatures in the measured dependences show that probably the dielectric and magnetic constants and dielectric and magnetic loss tangents decrease above 15 – 20 GHz (especially the parallel values $\varepsilon//\tan\delta//$). These parameters will be determined to a higher degree of accuracy as part of future works. The observed differences notwithstanding, we are satisfied with the results – to the best of our knowledge, for the first time we obtained the dielectric and magnetic parameters of a Z-type ferrite ($\mathrm{Sr}_3\mathrm{Co}_2\mathrm{Fe}_{24}\mathrm{O}_{41}$ hexaferrite in the case discussed here) in the microwave range above 3 GHz. The broadband method proposed – covered CPW and MSL structures, ensures a very important advantage: possibility to apply external dc magnetic fields in parallel and perpendicular directions to the disk sample. The insets in figure 4 show the configurations used. The experiments allowed us to ascertain the existence of the so-called magneto-electric (ME) effect in the multiferroic material considered. The first results are presented in figure 4 b; they definitely show that the hexaferrite investigated exhibited an ME effect – the phase delay increases for perpendicular external dc magnetic induction ~ 0.1 T (dielectric constant increases); while this delay decreases for parallel field (dielectric constant decreases). A similar behavior is manifested by the losses; they decrease for a parallel dc magnetic field and increase for a perpendicular one.

![Image 1](image1.png)

![Image 2](image2.png)

**Figure 4.** a) Additional losses and phase delay in MSL covered by SUT with parameters taken from the last row in table 1 (solid curves – measurements; dashed curves – simulations by ANSYS HFSS®); b) changes in the additional losses and phase due to magnetization of hexaferrite SUT in parallel and perpendicular direction – shown in the insets to the figures.

### 4. Conclusions and future work

To the best of our knowledge, we succeeded for the first time in measuring the dielectric and magnetic parameters of a Z-type hexaferrite ($\mathrm{Sr}_3\mathrm{Co}_2\mathrm{Fe}_{24}\mathrm{O}_{41}$) in the frequency range 5 – 40 GHz and established a measurable anisotropy of the permittivity and permeability. A change of both parameters was also observed (ME effects) in an external dc magnetic field applied in parallel and perpendicular directions.
to the sample at room temperature. The experiments conducted are complex so that the results are of moderate accuracy. They should be considered as a first step in the process of a comprehensive characterization of hexaferrite magnetoelectrics above 3 GHz up to the mm-wave range. We intend to deepen and widen this type of research (including ME effect) by performing accurate perturbation measurements on small and thinner samples (prisms and disks).

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References
[1] Harris V G 2012 IEEE Trans. on Magnetics 48/3 1075-1104
[2] Kimura T 2012 Magnetolectric Hexaferrites, Chapter in Annu. Rev. Condens. Matter Phys. 3 93-110
[3] Tang R, Jiang Ch, Qian W, Jian J, Zhang X, Wang H and Yang H 2015 Dielectric relaxation, resonance and scaling behaviors in Sr$_3$Co$_2$Fe$_{24}$O$_{41}$ hexaferrite Scientific Reports 5 13645 DOI: 10.1038/srep1364
[4] Ebnabbasi K, Chen Y, Geiler A, Harris V and Vittoria C 2012 J. Appl. Phys. 111 07C719(2012); doi: 10.1063/1.3678588
[5] Koutzarova T, Ghelev Ch, Peneva P, Georgieva B, Kolev S, Vertruyen B and Closset R 2018 J. Phys.: Conf. Series 992 012058
[6] Dankov P I 2006 IEEE Trans. on MTT 54/4 1534-1544 DOI: 10.1109/TMTT.2006.871247
[7] Dankov P 2017 Proc. iWAT’2017 DOI: 10.1109/iWAT.2017.7915316, IEEE Xplore online available
[8] Dankov P I 2010 Dielectric anisotropy of modern microwave substrates Ch. 4 in Microwave and Millimeter Wave Technologies from Photonic Bandgap Devices to Antenna and Applications Ed Minin I (In-Tech Publ.) ISBN 978-953-7619-66-4
[9] Dankov P, Tzaneva B and Videkov V 2019 Proc. SPIE 11332 Int. Conf. on Quantum, Non-linear and Nanophotonics (ICQNN 2019) 1133203 (30 Dec. 2019); doi: 10.1117/12.2551892