

Magnetolectric gradient structures: Properties and applications

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This paper is devoted to a comprehensive study on a new type of microwave structures named magnetolectric (ME) gradient structures. These structures are studied in this paper to understand the possibilities and application principles in feasible devices. The structure under study was calculated at different values of the applied electric field and different values of the relative permittivity of the artificial dielectric layer. The layered multiferroic structure in inhomogeneous electric and magnetic fields was calculated on the basis of the previously proposed mathematical model. The eigenwaves spectrum for several considered cases was the result of the performed calculation. The concept of using ME gradient structures in the design of electronically controlled microwave devices is formed on the basis of the results of a numerical experiment. Structures of this type will preferably be used in electronically controlled devices for the directional transmission of microwave signals, as it was shown in the theoretical part of the paper.

Keywords: Composite structures; multiferroics; magnetolectric gradient structures; artificial dielectric; eigenwave spectrum.

1. Introduction

Multiferroics as a special class of substances have been of great interest for research recently. The class of multiferroics with the presence of magnetolectric (ME) interaction, which consists of inducing magnetization when exposed to an external electric field on the material and vice versa inducing polarization when exposed to a magnetic field, deserves special attention.

The first stage of materials research in order to understand the interaction of ferromagnetic and ferroelectric phases dates back to the second half of the 20th century.¹ The magnitude of the ME effect at the time of the discovery of this interaction was insignificant for practical application, for this reason, further work of the first stage of research was focused on the search for materials with a greater magnitude of this effect. A new impetus in the study of ME materials was given by the transition to the use of layered composite structures. The first works in the field of practical application of the ME effect, including for use in ME controlled microwave devices, belong to the scientific research group of Bichurin.² The devices that were developed by this research group had the ability to control the electrical potential which gives a faster adjustment of characteristics and a significant reduction in control power. For example, in comparison with the traditional methods of controlling ferrite microwave devices by means of a magnetic field, it gives significant advantages.

The term ME gradient structures was first used in Ref. 3 to describe a new type of composite structures which are

the product of the integration of a ME, multiferroic and an artificial dielectric. Note that such combination will qualitatively expand the possibilities of electronic control of the wave properties of these structures, for example, to the generalized mechanism described in Ref. 4 by means of the control action of the electric field. This will add an artificial gradient distribution of the permittivity of one of the dielectric layers of the structure.

A comprehensive study of the properties of new type of structures from the point of view of the principles of their subsequent practical application should be the next step in the study of this type structures.

A complex analysis of the ME gradient structures and the spectrum of electromagnetic eigenwaves based on the description of the constituent parts of the composite and a numerical experiment on the base of previously constructed mathematical model in this study was investigated. The obtained theoretical data formed the basis for subsequent experimental studies in terms of the practical application of this type of structure.

2. Magnetolectric Gradient Structures

2.1. *Multiferroic layered ferroelectric-ferrite structures and effects in them*

The period of intensive and successful development of the magnetic phenomena physics and the ferroelectrics physics preceded the in-depth research of multiferroics. Two types of

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multiferroics are distinguished today. The first type contains materials in which the electric polarization is independent (or weakly coupled) from the magnetic order, i.e., materials in which the magnetic and ferroelectric properties are due to different sources. Ferroelectric properties are usually manifested in these materials at higher temperatures than magnetic ones and spontaneous polarization reaches rather large values ($\sim 10\text{--}100\text{ uC/cm}^{-2}$). Examples of the multiferroics of the first type are BiFeO_3 , YMnO_3 , etc.

The second type of multiferroics is magnetic multiferroics, in which electric polarization is generated by magnetic ordering. The materials of this group are characterized by a strong connection between magnetic and ferroelectric properties, but their spontaneous polarization is significantly less than that of the first type of multiferroics ($\sim 10^{-2}\text{ uC/cm}^{-2}$). Practically all multiferroics are synthesized compounds. Only two natural crystals are known, these are congolite $\text{Fe}_3\text{B}_7\text{O}_{13}\text{Cl}$ and chambersite $\text{Mn}_3\text{B}_7\text{O}_{13}\text{Cl}$.

Devices using multiferroics can be classified into three types. The first type is devices that use ferroelectric or magnetic properties separately. The second type is devices in which ferroelectric and magnetic properties are used simultaneously, but without ME interaction, for example, a Faraday phase shifter. The third type is such devices whose action is based on the ME effect (they use multiferroics of the second type). The interaction of electrical and magnetic subsystems in multiferroics can manifest itself in the form of a number of effects that underlie the operation of devices of the third type.

Historically, the insignificant magnitude of the ME effect in monocrystals, and therefore the great complication of their practical use, shifted the focus of most applied research to the field of ME composite materials. It is known that the composites output properties are determined by the properties of individual phases and their interaction, these are properties such as sum properties, proportional properties and multiply properties. ME multiferroic composites can be classified as substances with multiple properties.

ME composites are divided into three structural types based on the components connectivity,⁵ as shown in Fig. 1: 0 are single-phase particles suspended in a matrix of another phase, which is indicated by the number 3; 1 are single-phase fibers, 2 are films or layers.

The combination of ferroelectric and ferrite materials made it possible to implement electronic adjustment by using two effects in the design of controlled microwave devices.

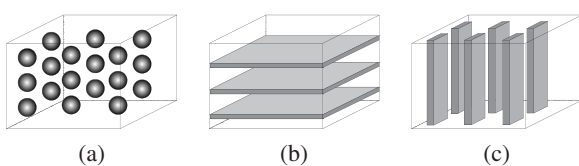


Fig. 1. Structural types of ME composites: (a) Composite of 3-0 type; (b) composite of 2-2 type and (c) composite of 1-3 type.

One of these effects is the reversal ME effect, which is a so-called “secondary” effect in the chain of transformations “piezoelectric effect–elastic deformation–change in magnetic anisotropy–shift of the ferromagnetic resonance (FMR) line”. In this case, it appears in the shift of the FMR line under the action of a constant electric potential. In this case, we deal with the reverse ME effect in the FMR region, which is of practical interest in the microwave range. The devices type in which the ME composite material acts as an active element is called the ME type of devices.⁶

The second effect that appears in ME composites and makes a significant contribution is the electrodynamic effect. It is caused by the interaction of electromagnetic waves in the ferroelectric layer and spin waves in the ferrite layer of the composite structure. A change in the permittivity of the ferroelectric layer under the action of an applied electric field leads to a change in the dispersion characteristics of the electromagnetic wave in this layer, and also leads to a change in the wave properties of the layered structure as a whole.

Mutual hybrid electromagnetic-spin waves propagate in layered ferrite-ferroelectric structures, as it was shown in Refs. 7 and 8. Such waves combine the properties of propagating waves, for example, in ferroelectric dielectric layers, they will be electromagnetic waves and in the ferrite layer — spin ones. This theoretical model was used in the development of phase shifters and resonators with implemented electric and magnetic control.^{9,10}

Microwave devices based on the ME effect, such as a receiving microstrip antenna,^{11,12} a filter-preselector,¹³ an attenuator,¹⁴ a phase shifter^{2,15} and a gyrator¹⁶ have also been well studied.

2.2. Artificial dielectric

A significant section in the composite media theory is assigned to artificial dielectrics (AD), which are artificially created by metal-dielectric structures. AD is a fully functional dielectric type used in practical applications. ADs are structurally similar to ME composites, but they are a set of metal particles interspersed with a given spatial distribution in a dielectric medium,¹⁷ as shown in Fig. 2.

The charges on each of the conducting plates are displaced by an external electric field, thereby simulating the behavior of dipoles in a conventional dielectric.¹⁸ The AD design can be identified in the first approximation with a homogeneous continuous medium (homogenization) with some effective

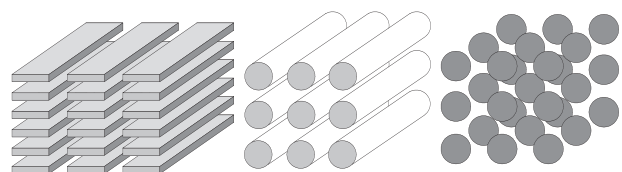


Fig. 2. Fragments of periodic sequences of metal particles of AD.

material parameters for the case when the electromagnetic wavelength exceeds of the metal inclusions location period.

ADs have found their application in microwave technology for manufacturing substrates in which an artificial increase in the dielectric constant allowed us to proceed to the design of compact microwave resonators.¹⁹ Also, AD substrates have found their application in the design of antennas,²⁰ high-impedance surfaces and anti-reflective coatings.²¹ The possibility of gradient distribution of dielectric permittivity in the AD layer is another advantage of the artificial medium.

2.3. Structure artificial dielectric–magnetolectric multiferroic

The supposed advantages of this type of layer merging were based on the following considerations. ME composite has a number of special properties in the microwave region. The dependence of ME material characteristics in the FMR region on the values of stationary electric and magnetic fields⁶ and the dependence of the permittivity for ferroelectric appears on the value of the stationary electric field²² are the most important. Each of these properties contribute to an important characteristic of ME devices, namely electronic control of the dispersion characteristics of their eigenwaves through changes in stationary electric and magnetic fields.

These dependences can be described mathematically using the thermodynamic description of multiferroic composites through the differential expression of Gibbs free energy²³:

$$dG = -SdT - xd\sigma - PdE - MdH, \quad (1)$$

where S is the entropy,
 T is the temperature,
 x is the strain,
 σ is the external applied mechanical stress,
 P is the polarization,
 M is the magnetization,
 E and H are applied external electric and magnetic fields, respectively.

Expression (1) for the case of an isothermal adiabatic system will have the following form:

$$\begin{cases} x = s\sigma + d^e E + d^m H \\ P = d^{e,c} \sigma + \chi^e E + \alpha^m H, \\ M = d^{m,c} \sigma + \alpha^e H + \chi^m H \end{cases}, \quad (2)$$

where d^e , $d^{e,c}$ и d^m and $d^{m,c}$ are direct and inverse piezoelectric and magnetostrictive coefficients, respectively,

$\chi^{e,m}$ is the dielectric and magnetic susceptibility,
 $\alpha^{e,m}$ is the ME coefficient.

The ME composite magnetization and polarization and consequently the frequency spectrum of the eigenwaves of the entire structure depends on the magnitudes of the applied fields, as shown by the system of expressions (2). Note,

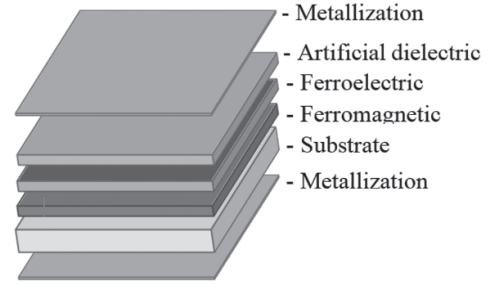


Fig. 3. The artificial dielectric–magnetolectric multiferroic structure.

however, that the entire structure is subjected to such an influence.

A built-in control system for local or point parameters inside the structure by means of electric and magnetic fields excited by point sources could be given as an example of solving the problem of local action. However, this method of controlling the parameters of the material structure is technically difficult and inefficient, and this creates a problem for managing the parameters of such an environment.

ME structures with an integrated artificial dielectric can solve this problem (see Fig. 3).

The integration of an additional dielectric layer into the structure of the composite leads to a change in the permittivity of the entire structure, therefore, the frequency spectrum also changes. The possibility of gradient designing of the dielectric constant of an additional integrated layer exists at the design stage. In this case, it is possible to assign a different response for the same applied field to different areas of the same structure. Consequently, a new property of controlled local changes in the microwave properties of the structure appears through gradient designing of the dielectric constant.

3. Applications of Magnetolectric Gradient Structures

3.1. Eigenwave spectrum of the ME gradient structure

A four-layer structure in which layers 2–4 are the ME multiferroic area and layer 1 is set as the AD which will be considered for mathematical confirmation of the above as shown in Fig. 4.

This problem from the point of view of Maxwell’s electrodynamics was analyzed in detail in Ref. 3. The dispersion equation into which the dependencies were introduced was obtained as a result: The dependences of the permittivity on the electric field were obtained for the ferroelectric layer $\varepsilon_2(E)$; dependences of the effective internal magnetic field on the electric field were obtained for the ferrite layer $\omega_H = \gamma H_{eff}(E)$.

The following parameters were used to calculate the dispersion characteristics of the structure shown in Fig. 4: Layer 1 is AD with thickness of $d_1 = 100 \mu\text{m}$; layer 2 is ferroelectric

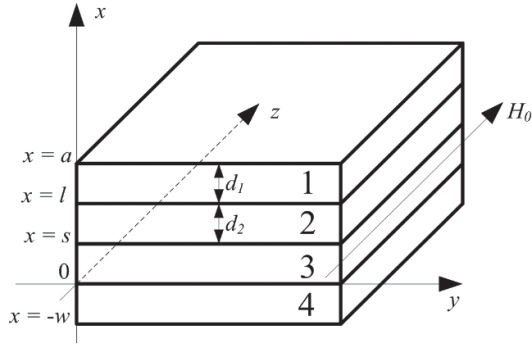


Fig. 4. ME gradient structures.

lead zirconate titanate (PZT) with thickness of d_2 equal to 100 μm , the relative permittivity is $\varepsilon_2(0)$ equal to 1870; ferromagnetic layer 3 is yttrium iron garnet (YIG) with thickness s equal about 5 μm , saturation magnetization is M_0 about 1750 G, external constant magnetic bias field of ferromagnetic is H_{e0} about 2022 Oe; dielectric layer 4 is gadolinium gallium garnet (GGG) with thickness of w equal to 500 μm , the relative permittivity is ε_4 of 11. The calculation was carried out for two values of the relative permittivity of an AD: (a) $\varepsilon_1 = \varepsilon_2(0) = 1870$, the relative permittivity AD is equal to the relative permittivity of the ferroelectric at zero applied electric field; (b) $\varepsilon_1 = 3400$. In addition, for two values of the applied stationary electric field: 0 kV/cm and 40 kV/cm. The results are presented in Fig. 5.

3.2. Discussion of the results

The electric field, which affects the structure under study, leads to a change in the spectrum of its eigenwaves in it, i.e., a growth in the frequency of dispersion branches, as can be seen from the obtained dispersion characteristics which is shown in Fig. 5(a). This is due to a decrease in the dielectric permittivity of the ferroelectric in particular and a decrease of the total dielectric permittivity of the upper layers in general for the AD–ferroelectric layers. On the other hand, an increase in the permittivity of an AD leads to a frequency decrease in the dispersion branches (see Fig. 5(b)). That is, situations in which these two processes will acquire relative equality may be possible. This was achieved in the calculation example

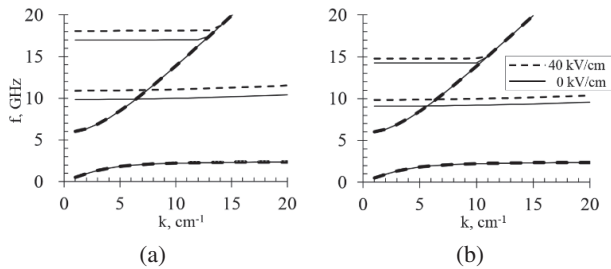


Fig. 5. Spectrum of eigenwaves of the structure under study: (a) $\varepsilon_1 = \varepsilon_2(0) = 1870$; (b) $\varepsilon_1 = 3400$.

shown in Fig. 5, where equality was found by selecting the values of the applied electric field and the AD permittivity: The spectrum of eigenwaves at frequencies up to 12–14 GHz at zero electric field and relative permittivity AD $\varepsilon_1 = \varepsilon_2 = 1870$ practically coincided with the spectrum at $E = 40$ kV/cm and value of $\varepsilon_1 = 3400$.

3.3. Principles of application of the ME gradient structures

Suppose that the ME gradient structure under consideration consists of four layers similar to the one under study (Fig. 4), but has two regions in which the values of the relative permittivity of the layer with an AD have the values $\varepsilon_1 = 1870$ and $\varepsilon_1 = 3400$. The symbols **A** and **B** denote the corresponding areas, and y is the direction of wave propagation, as shown in Fig. 6.

The spectrum of eigenwaves in each of the regions of the structure under consideration will have the form shown in Fig. 5. Some simplifications will be accepted for calculations: the spectral region of the structure **A** is preassigned, as well as the assumption that an electromagnetic wave passing through this structure obtain a preassigned phase shift. The applied electric field gives the effect of the shift of the initial spectral region. In other words, the frequency spectrum of region **B**, if an external electric field is applied to it, becomes similar to the spectrum of region **A** with zero external electric field. As a result, the ability to form a predefined, electronically controlled, phase front at the output of the entire structure by selecting the values of the dielectric constant of the AD and of the applied electric field magnitude appears.

The following example of the use of ME gradient structures is a special case based on the experimentally revealed phenomenon of the transformation of surface magnetostatic waves into electromagnetic waves by scientists Vashkovskij and Lock (MSSW-to-EMW transformation).^{24–26} The general meaning of the phenomenon is as follows: If the wavelength increases as the MSSW propagates in inhomogeneously magnetized films of YIG, then in the region of the film surface, where the wave number of MSSW becomes close to the wave number of EMW of the adjacent space, the MSSW-to-EMW transformation one take place (see Fig. 7(a)). Power conversion up to 90% was recorded in experimental work. The addition of a ferroelectric layer to the previous structure makes it possible to control the MSSW-to-EMW conversion

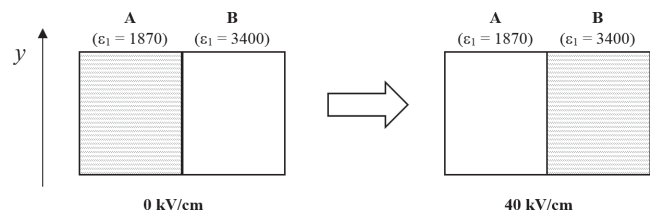


Fig. 6. Principles of application ME gradient structures.

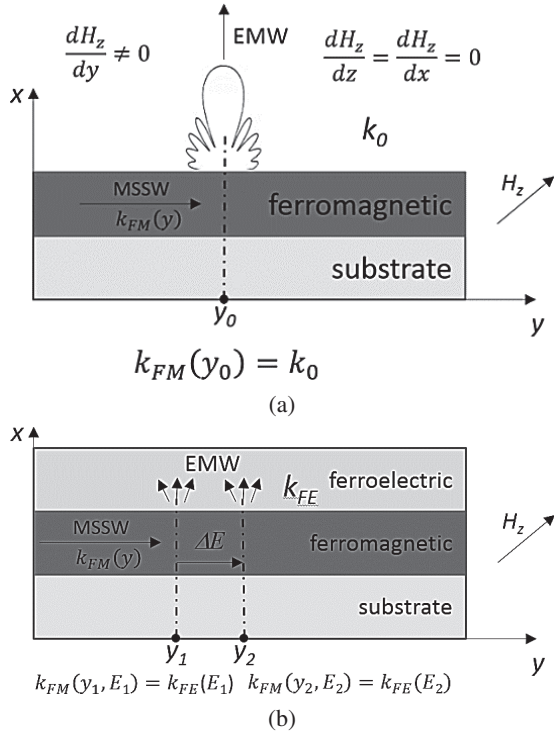


Fig. 7. Graphical representation of the MSSW-to-EMW transformation process: (a) Into free space and (b) in the case of ME composite.

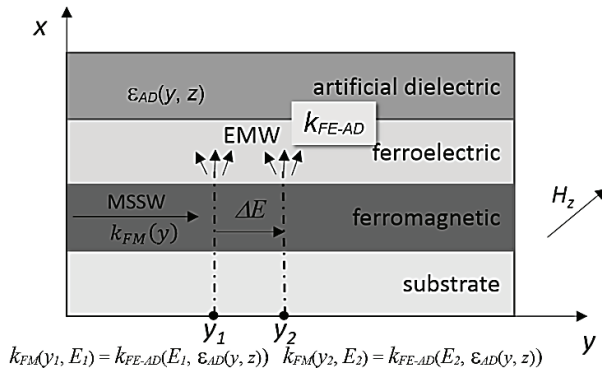


Fig. 8. Graphical representation of the MSSW-to-EMW transformation process for an ME gradient structure.

by means of an electric field E through the ME effect and the effect of the dependence of the dielectric permittivity of the ferroelectric on the magnitude of the applied electric field (see Fig. 7(b)).

The areas of MSSW-to-EMW transformation can be set locally by adding an additional layer of the AD to the multiferroic structure (by artificial distribution the permittivity), as it shown in Fig. 8.

4. Conclusion

ME gradient structures are a new and promising type of electronically controlled microwave structures. The new

structures proposed by us structurally represent a higher-level composite, the components of which are the ME multiferroic and the AD also are composite materials. ME gradient structures possess the properties of their components and expand the range of control of microwave devices developed on their basis in comparison with the original prototypes. In devices based on the proposed structures, in addition to the electronic control of the ME of microwave devices that have been studied in sufficient detail, a new way is added to control the properties of structures and thus microwave devices via an artificially set distribution of the permittivity of an additional dielectric layer.

The paper is devoted to a comprehensive study four-layer layered structure consisting of an artificial dielectric and an ME multiferroic was studied based on the previously proposed mathematical model. The microwave spectrum of eigenwaves was obtained by the calculation method for two values of the dielectric permittivity of the AD layer and two values of the amplitude of the applied electric field. The effect of increasing the value of the dielectric constant of the AD layer is the opposite of the effect of applying an electric field as it was shown in the calculations. Computations also show that a situation of balance of these effects is possible, which lays the foundation for the principles of designing devices based on the structure under study.

As it follows from the current state of the proposed technology, in the near future, ME gradient structures are most preferably used in the development of solid-state microwave electronically controlled devices for directional energy transmission: Directional couplers, circulators, valves and patch antennas.

In conclusion, it should be noted that the possible dielectric losses in the ME gradient structure also needs to be calculated to understand the full picture of the propagation of the electromagnetic waves. However, such a study requires separate consideration based on both theory and experiment.

The ME gradient structure is a complex layered composite and therefore the dielectric losses will be determined by different sources. It is known that the dielectric losses are proportional to the square of the applied electric field and the frequency. Ferroelectric as part of the structure under consideration will be a source of losses associated with spontaneous polarization. These losses are significant at temperatures below the Curie point. Dielectric losses due to through conductivity and heterogeneity of the structure will be characteristic of the AD. The latter dependence is difficult to theoretically analyze and can be determined experimentally, which is important for understanding.

Based on the above, several recommendations can be given to reduce the influence of these phenomena on dielectric losses in devices based on ME gradient structures. Firstly, ferroelectrics with a narrow polarization hysteresis loop, i.e., low values of residual polarization and coercive fields can be used in the proposed applications. It is worth pointing out that barium–strontium titanate (BST)^{9,10,27} is promising for

use for ME gradient structures. Second, metal particles of a disk shape can be used to avoid the effect of through conduction in the AD (see Ref. 20).

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