Functional Materials 21, No.3 (2014)

Magnetoresonance properties of manganite-perovskite nanocomposite with Bi2223-superconductor additives at $T = 300$ K, 4.2 K

T.V.Kalmykova, S.Yu.Polevoy, S.V.Nedukh, S.I.Tarapov, V.Yu.Tarenkov*

Institute of Radiophysics and Electronics, National Academy of Sciences of Ukraine, 12 Acad. Proskura St., 61085 Kharkiv, Ukraine
*Donetsk Physics & Technology Institute, National Academy of Sciences of Ukraine, 72 R.Luxemburg St., 83114 Donetsk, Ukraine

Received February 10, 2014

Results of research of electron magnetic resonance absorption in nanocomposites with different ratios of $La_{0.6}Sr_{0.4}MnO_3$ manganite and superconductor $Bi_{1.7}Pb_{0.3}Ca_2Sr_2Cu_3O_{7-x}$ (Bi2223) additives in the wide range of frequencies of 22–40 GHz, 68–80 GHz are presented at temperatures $T = 340$ K and 4.2 K. The manifestation of two peaks in the Electronic Magnetic Resonance spectrum is assigned to appearance of collinear ferromagnetic and paramagnetic phases in the structure under study.

Представлены результаты исследования магниторезонансного поглощения электромагнитных волн в нанокомпозитах с разным соотношением манганита $La_{0.6}Sr_{0.4}MnO_3$ и сверхпроводника $Bi_{1.7}Pb_{0.3}Ca_2Sr_2Cu_3O_{7-x}$ (Bi2223) в широком диапазоне частот 22–40 ГГц, 68–80 ГГц при комнатной и гелиевой температуре. Продемонстрировано наличие двух пиков электронного магнитного резонанса, приписываемых коллинеарной ферромагнитной и парамагнитной фазам.

Магніторезонансні властивості нанокомпозиту манганіту-перовскіту з додаванням надпровідника Bi2223 при $T = 300$ К, $T = 4.2$ К. *Т.В.Калмикова, С.В.Недух, С.Ю.Полевой, С.І.Тарапов, В.Ю.Таренков.*

Представлено результати дослідження магніторезонансного поглинання електромагнітних хвиль у нанокомпозитах з різним співвідношенням манганіту $La_{0.6}Sr_{0.4}MnO_3$ та надпровідника $Bi_{1.7}Pb_{0.3}Ca_2Sr_2Cu_3O_{7-x}$ (Bi2223) у широкому діапазоні частот 22–40 ГГц, 68–80 ГГц при кімнатній і гелієвій температурі. Продемонстровано наявність двох піків електронного магнітного резонансу, що належать колінеарній феромагнітній і парамагнітній фазам.

1. Introduction

Interest to research of magnetic materials such as, for example, manganites perovskites is caused both the demands of modern technologies and abundance of various types of the magnetic interactions in the crystal lattice of perovskite, called by their intrinsic features. One of the most prominent features of such structures is the effect of Colossal Magnetoresistance (CMR) which for manganites at cryogenic temperatures can extend in some orders similar effect for traditional multilayered heterostructures.

Materials with general formula $Ln_{1-x}A_xMnO_3$ (where Ln is a trivalent ion of group La, A is a bivalent ion of alkaline or alkaline-earth atom) are of strong interest due to their complex phase diagram. The main mechanism of the charge transfer in such structures is the "double exchange" mechanism between Mn-ions of different valency (Mn^{3+}–O–Mn^{4+}) [1]. The "double exchange" and Yan-Teller effect allow to explain low-temperature properties of weakly doped compounds of $LaMnO_3$ in the frame of model of band insulator model.

Another important fact is that the doped manganites-perovskites can reveal the left-handed properties [2–4] in the microwave band. Existence of these properties and opportunity to control them by temperature and by external magnetic field also causes a great interest from the practical and fundamental points of view. Otherwise it is known [5, 6], that in nanocomposite materials of various consistence such as: superconductor-magnetic, metal-dielectric material, etc. very specific mechanisms of transfer of a charge and dynamics of a magnetic flux can arise, that can affect on the electromagnetic features of structures under research. Namely by this reason the magnetic properties of manganite-perovskite with addition of high-temperature superconductor $Bi_{1.7}Pb_{0.3}Ca_2Sr_2Cu_3O_{7-x}$ (the ceramics of bismuthic metaloxide which abbreviation is Bi2223) are investigated in the given work.

2. Experimental

We studied the electron magnetic resonance response for three samples of $La_{0.6}Sr_{0.4}MnO_3$ + Bi2223 manganite perovskite, in which Bi2223 concentration was 85 %, 79.5 %, 75 %, correspondingly, at temperatures $T = 4.2$ K (frequencies $f = 60$–78 GHz) and $T = 300$ K (frequencies $f = 2$–40 GHz).

The ceramics of bismuthic metaloxide of the phase Bi2223 was obtained from the corresponding oxides by the method of solid-phase synthesis at temperature 850°C. After secondary re-grinding and repressing samples were annealed in 2 stages: at the first stage annealing was carried out at $T = 850$°C for 8 h and at the second stage annealing was carried out at $T = 825$°C for 6 h. Such technology allows to receive the concentration of Bi2223 in the sample up to 95 %. Electronic microscopic researches showed, that the samples represent the ceramic polycrystal consisting of well-expressed microcrystals with $d = 5$–10 μm sizes.

Manganite perovskite specimens $La_{0.7}Sr_{0.3}MnO_3$ having sizes of nanoparticles of $d = 8$–10 nm were obtained by Zol-Gel method. On the basis of these components the composite with various volume contents of component was obtained. To reach the maximum homogeneity of the specimen the powders of Bi2223 and manganite — $La_{0.7}Sr_{0.3}MnO_3$ were mixed in alcohol, dried and stirred mechanically. After that the tablets (with diameter $d = 18$ mm) were pressed at $P = 10$ Kbar. To verify the technology the plate shaped specimens $S = 0.2 \times 1 \times 10$ mm^3 sizes were performed simultaneously. Current and potential contacts were prepared by drawing the colloidal silver into the contact area before sample pressing. The samples weren't sintered to avoid the interdiffusion and chemical reactions between the components.

Magnetic resonance absorption in the range of 22–40 GHz; (T = 300 K)

Experimental registration of magnetoresonance absorption has been carried out using Vector Network Analyzer Agilent PNA-LN5230A by the technique described in [7]. Studied samples during experiment were placed in a metal rectangular waveguide which was located in turn in a gap between electromagnet poles. The specimens: No.1 — $La_{0.6}Sr_{0.4}MnO_3$(15 %) + Bi2223 (85%), No.2 — $La_{0.6}Sr_{0.4}MnO_3$(20.5 %) + Bi2223 (79.%), No.3 — $La_{0.6}Sr_{0.4}MnO_3$(25 %) + Bi2223 (Bi 75 %) have been studied.

During experiment the magnitudes of halfwidth of the Electron Resonace Line (the linewidth, $\Delta H_{1/2}$) were determined. For "parallel" geometry of experiment, (the vector of external magnetic field and magnetic component of the microwave field vector lie in the plane of the thin-film sample and are mutually perpendicular) it was $\Delta H_{1/2}$ = 2000–2500 Oe. For "perpendicular" geometry, (microwave field magnetic component lies in the sample plane, and the vector of magnetic field is directed perpendicularly to the sample plane) it was $\Delta H_{1/2} = 3000$ Oe. The resonance frequency magnitudes versus the resonance magnetic field magnitudes dependencies (called as resonance frequency-field dependences) are presented in Fig. 1(a–c).

In Fig. 1 the dotted line is the reference curve representing the known resonance frequency-field dependence for free electron, by squares — the resonance frequency-field dependence for "parallel" geometry of the experiment, triangles — resonance frequency-field dependence for "perpendicular" geometry of the experiment.

Magnetic resonance absorption in the range of 60–78 GHz; (T = 4.2 K)

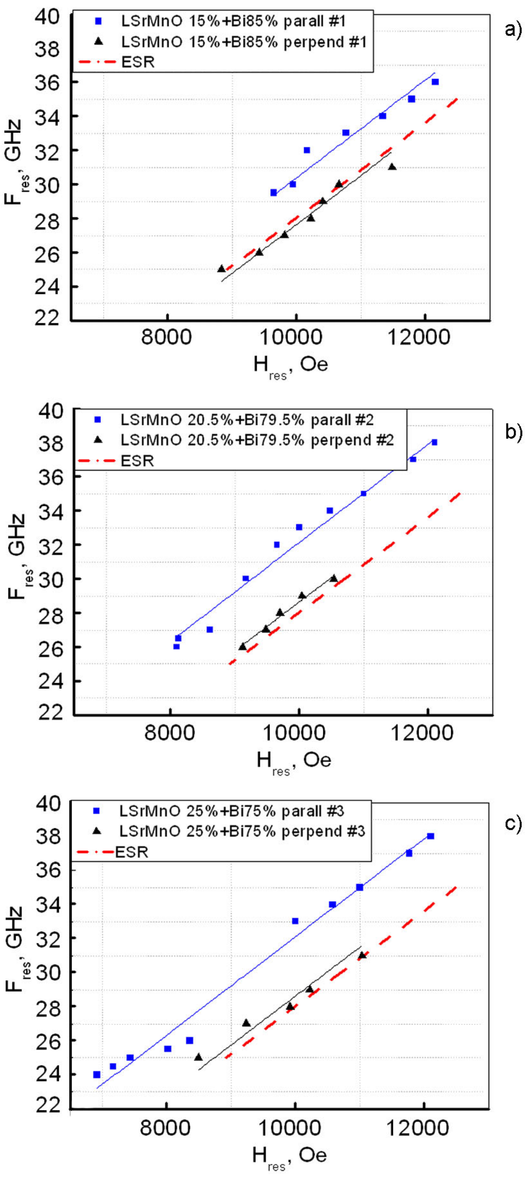

Fig. 1. Resonance frequency-field dependence at T=300 K: a) №1 $La_{0,6}Sr_{0,4}MnO_3$(15%) + Bi2223(85%), b) №2 $La_{0,6}Sr_{0,4}MnO_3$(20.5%)+ Bi2223(79.5%), c) № 3 $La_{0,6}Sr_{0,4}MnO_3$(25%)+ Bi2223(Bi75%). Squares – the "parallel" geometry; triangles – the "perpendicular" geometry; dashed line – the reference curve.

Cryomagnetic radiospectrometer BURAN [8] was applied to carry out the low-temperature magnetic resonance experiment.

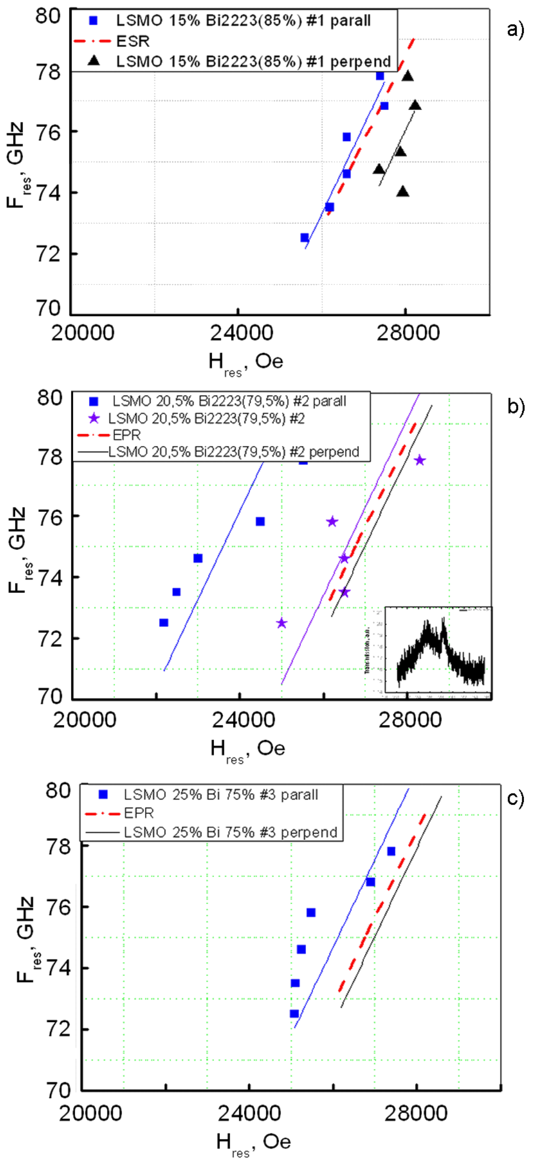

Fig. 2 Resonance frequency-field dependence at T=4.2 K a) № 1 $La_{0,6}Sr_{0,4}MnO_3$(15%)+ Bi2223(85%), б) № 2 $La_{0,6}Sr_{0,4}MnO_3$(20.5%)+ Bi2223(79.5%), в) № 3 $La_{0,6}Sr_{0,4}MnO_3$(25%)+ Bi2223(Bi75%). Squares – the "parallel" geometry; triangles – the "perpendicular" geometry; dashed line – the reference curve; stars – the satellite peak.

In Fig. 2(a–c) (resonance frequency)-(resonance field) dependences for three mentioned above samples, are presented for T = 4.2 K. The dependencies are based on the experimentally detected peaks of Electron Magnetic Resonance. Here the dotted line is the reference curve, represented the known resonance frequency-field dependence for not coupled free electron, squares — the resonance frequency-field dependence for "parallel" geometry of the experiment, triangles — the resonance frequency-field dependence "perpendicular" geometry of the experiment.

The graph with splitted line of the Electron Magnetic Resonance was detected for "parallel" geometry (see the inset in Fig. 2b). On the resonance frequency-field depend-

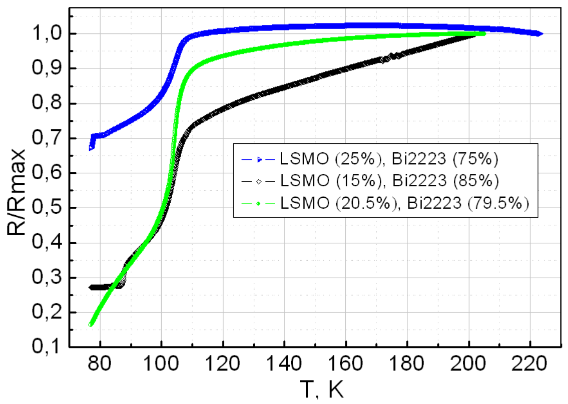

Fig.3 Temperature dependences of R/Rmax(T) for samples of Bi2223 – LSMO with volumetric concentration of 15, 20.5 and 25% of LSMO.

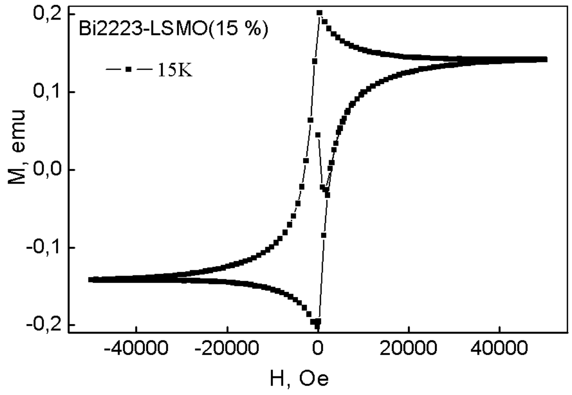

Fig. 4 M(H) dependence of a composite Bi 2223 - LSMO with volumetric concentration 15 % LSMO.

ence these points are marked by squares (the main peak) and stars (the satellite peak). This result is similar to result of work [9] where it was shown how for $La_{1-x}Bi_xMnO_{3+\delta}$ sample the line of the Ferromagnetic Resonance separates from the line of the Electron Spin Resonance while the temperature falls down.

4. Results and discussion

It is necessary to take into account that the samples under study are in two different "magnetic phase" states for various temperatures of the experiment. At temperature $T = 300$ K the LSMO component of the samples is ferromagnetic conductor and Bi2223 component still not superconductor. At $T = 4.2$ K the Bi2223 component is diamagnetic superconductor (Fig. 3 and 4), as the LSMO part is the most probably a mixture of ferromagnetic and superparamagnetic conductive phases.

Let analyze the behavior of the samples at temperature of $T = 4.2$ K, that is of the most interest.

In Fig. 3 $R/R_{MAX}(T)$ dependencies for the samples with 15, 20.5, 25 volume % of LSMO are presented. With increasing of the LSMO concentration the specific resistance of a composite grows, and $R/R_{MAX}(T)$ characteristic around superconducting transition of Bi2223 is washed away and at 77 K resistance of the samples becomes significantly large magnitude. This result specifies that the percolation cluster of Bi2223 even at small concentration of LSMO is broken. This fact contradicts conclusions of the classical theory of percolation which says that the infinite percolation cluster remains even to 80 % of impurity. Explanation for the fast breaking the percolation cluster of Bi2223 could be different dimensions of component, forming the composite. If Bi2223 has characteristic sizes of granules 5–10 μm, the LSMO consists of nanogranules of 8–10 nm. Most likely, nanogranules of manganite "flow round" the macroscopic granules of Bi2223 and the measuring current flows on Bi2223–LSMO–Bi2223–LSMO–Bi2223 chains.

General characteristic describing the superconducting and magnetic properties at low temperatures ($T = 15$ K) is $M(H)$ curve. As we can see, in Fig. 4 the dependence $M(H)$ presented has incomes from the both magnetic and diamagnetic components.

Presence of the LSMO ferromagnetic component leads to hysteresis shape of the loop of the magnetization reversal and the diamagnetic contribution Bi2223 (which in superconducting state) is manifested in the curving up parts of the magnetization reversal curve during the reversal path of magnetic field.

It testifies that the studied composite possesses properties of the both superconductor and ferromagnetic.

This assumption explains the results of the magnetoresonance experiments. Namely, the magnetic resonance absorption peak broadens with the temperature decreasing: from $\Delta H_{1/2} = 2000–2500$ Oe at $T = 300$ K up to $\Delta H_{1/2} = 3000$ Oe at $T = 4.2$ K. Beside this not one but two magnetoresonance peaks (Fig. 2b) present for the sample No.2. Thus while the temperature decreases up to range $T < T_B$, (here T_B — is the blocking temperature [10, 11]), a certain part of LSMO granules in the samples transfer into superparamagnetic state. This causes the appearance of additional magentoresonance peaks, which corresponds to superparamagnetic phase. Absence of the additional (satellite) magnetoresonance peaks for the samples No.1 and No.3, apparently is caused the fact that very broad magnetoresonance absorption peaks for ferromagnetic phase hide quite narrow absorption peaks of superparamagnetic LSMO granules.

Analysis of the results of the electron magnetic resonance absorption at T = 300 K is not so interesting because at such high temperatures the specific features of Bi2223 components in the composite are lost (which is normal but not in superconducting state). However, a qualitative analysis of these curves indicates that the samples of LSMO component are in ferromagnetic state and does not exclude the presence of the magnetic anisotropy field, directed perpendicular to the plane of the flat samples.

4. Conclusions

Thus, in the magnetic resonance researches of $La_{0.6}Sr_{0.4}MnO_3$ + Bi2223 nanocomposite: it was shown that the Electron Magnetic Resonance peaks for all samples broadened out when temperature fell up to T = 4.2 K. In sample No.2 at T = 4.2 K for "parallel" geometry, two lines of the Electron Magnetic Resonance registered, that testifies the co-existence of two magnetic phases: the ferromagnetic and superparamagnetic phase. It was shown that at temperature T = 15 K the Bi2223-component in sample No.1 is in the superconducting state.

Work was partially supported by the grant of Program of NAN Ukraine "Fundamental problems of nanostructural systems, nanomaterials, nanotechnologies", STCU#5714 grant and the Youth Scientists NASU Grant No.10/13.

References

1. K.Dorr, *J. Phys. D:Appl. Phys.*, **39**, R125 (2006).
2. A.Pimenov, A.Loidl, P.Przyslupski, B.Dabrowski, *Phys.Rev. Lett.*, **95**, 247009 (2005).
3. S.I.Tarapov, D.P.Belozorov, *Low Temper. Phys.*, **38**, 766 (2012).
4. D.P.Belozorov, S.I.Tarapov, A.M.Pogorily et al., *Appl. Phys. Lett.*, **100**, 171104 (2012).
5. M.Z.Meilikhov, *J.Exper. and Theor. Phys.*, **88**, 819 (1999).
6. R.V.Vovk, N.R.Vovk, O.V.Shekhovtsov et al., *Superconductor Sci. and Techn.*, **26**, 085017 (2013)
7. T.V.Kalmykova, S.I.Tarapov, S.V.Nedukh et al., *Functional Materials*, **19**, 14 (2012).
8. S.Tarapov, Gebze: Publ. Center of Gebze Institute of Technology (2000), p.93.
9. A.M.Pogorily, A.I.Tovstolytkin, D.M.Polishchuk et al., Proc. of MSMW'13, Ukraine (2013), p.23.
10. J.L.Dormann, D.Fiorani, E.Troncc, *JMMM*, **202**, 251 (1999).
11. Masatsugu Suzuki, Sharbani I.Fullem, Itsuko S.Suzuki et al., *Phys. Rev. B*, **79**, 024418 (2009).