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G. Li

G. G. Hu

H. D. Zhou

Xiaojuan Fan
Marshall University, fan2@marshall.edu

X. G. Li

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G. Li
Structure Research Laboratory, Department of Materials Science and Engineering, University of Science and Technology of China, Hefei 230026, People's Republic of China

G.-G. Hu
Department of Physics, Anhui University, Hefei 230039, People's Republic of China

H.-D. Zhou, X.-J. Fan, and X.-G. Li[a)]
Structure Research Laboratory, Department of Materials Science and Engineering, University of Science and Technology of China, Hefei 230026, People's Republic of China

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We present the frequency dependence of microwave-absorbing properties of $La_{1-x}Sr_xMnO_3$ ($x=0.4$, 0.5, 0.6, and 0.7) powders at room temperature. The absorbing properties change gradually with x in the frequency range of 8–12 GHz. The optimal absorption can be achieved for a $x=0.4$ sample and its microwave loss peak value is about 25 dB. Further experimental results show that the absorption can be attributed to magnetic and dielectric losses and the microwave loss peak corresponds to the maximum dielectric loss tangent $\tan\delta_e$ near 10.5 GHz. Furthermore, the absorbing properties of the oxides mixed with carbonic fiber and Y-type planar hexagonal ferrite have been rudimentarily studied. Results show that these additives greatly enhance the microwave-absorbing properties of the oxides.
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I. INTRODUCTION

With the development of radar, microwave communication technology, and especially the need for anti-electromagnetic interference coatings, self-concealing technology, and microwave darkrooms, the study of electromagnetic wave absorbing materials has increased in recent years.[1–3] The W-type and Y-type planar hexagonal ferrites are special kinds of absorbing materials due to their dielectric and magnetic losses in microwave frequency band. Usually, the resistivity of the ferrites is very high. However, the magnetic loss of the materials results from their ferrimagnetism, i.e., the resonance absorption of the moving magnetic domains and spin relaxation in the high frequency alternating electromagnetic fields.[4]

The distorted ABO_3-type perovskite oxides with the form of $RE_{1-x}AE_xMnO_3$ (RE=trivalent rare-earth elements; AE=divalent elements like Ca, Sr, Ba, and Pb) have been the subject of extensive investigations[5] because of the so-called colossal magnetoresistance effect and their potential applications in the magnetic recording industry. From the application viewpoint, one hopes that the materials have a higher Curie temperature, T_C (T_C is higher than the room temperature), and a larger magnetoresistance ratio in a low magnetic field. A $La_{1-x}Sr_xMnO_3$ ($x=0.4$, 0.5, 0.6, and 0.7) system[6,7] exhibits strong magnetism and low resistivity at room temperature suggesting that the system also has microwave-absorbing properties. Some authors[8–13] have reported the microwave losses, especially the microwave magneto-impedance in microsize powders of $La_{1-x}(Ca, Sr, Ba)_xMnO_3$ systems. It implies that the oxides have several useful applications such as acoustic modulators, magnetic field sensors, and microwave absorbers.

In this article, we reported the microwave-absorbing properties of $La_{1-x}Sr_xMnO_3$ ($x=0.4$, 0.5, 0.6, and 0.7) manganese oxides at room temperature in a zero magnetic field and studied the effects of additives like carbonic fiber and Y-type planar hexagonal ferrite on the absorption properties in the frequency range of 8–12 GHz.

II. EXPERIMENTS

The samples with the nominal composition $La_{1-x}Sr_xMnO_3$ ($x=0.4$, 0.5, 0.6, and 0.7) were prepared by a solid-state reaction method from the starting materials La_2O_3, $MnCO_3$, and $SrCO_3$ of high purity (>99%). The starting materials were first completely mixed, ground, and calcined at 1000 °C for 24 h leading to powders. Then, the powders were pressed into pellets and sintered at 1380 °C for 24 h twice in the air with intermediate grindings. Finally, the pellets were reground to powders with ~1 μm diameter particle size. X-ray diffraction reveals that the sintered samples have single phase with cubic perovskite structures.

The prepared powders were then mixed with wax by the ratio of 8:3 in weight to produce rectangular coatings of different thicknesses and attached to a square aluminum plate. The prepared coatings were inserted into the rectangular waveguide for measuring the microwave-absorbing properties and the complex magnetic permeability $\tilde{\mu}$ and dielectric permittivity $\tilde{\varepsilon}$. The magnetic and dielectric loss tangents, $\tan\delta_m$ and $\tan\delta_e$, were checked by measuring the coefficients of reflected and transmitted waves. In order to extract

[a)]Electronic mail: lixg@ustc.edu.cn

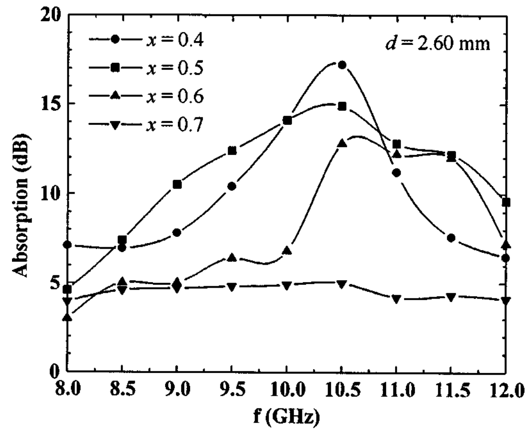

FIG. 1. The frequency dependence of the microwave absorption for $La_{1-x}Sr_xMnO_3$ ($x=0.4$, 0.5, 0.6, and 0.7) coatings with thickness of 2.60 mm.

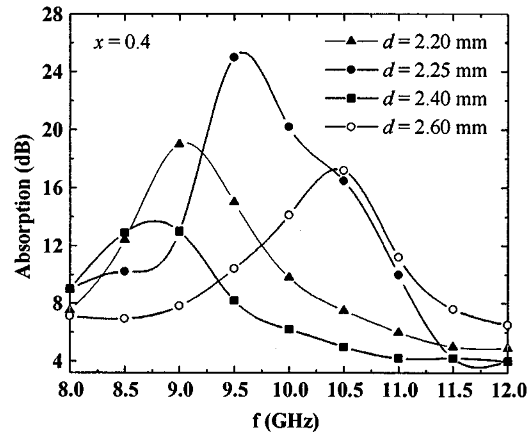

FIG. 2. The microwave absorption versus frequency for $x=0.4$ coatings with different thicknesses.

tan δ_e of the powders from the prepared coatings, the contributions of the uncoated aluminum plates to dielectric losses in the frequency range of 8–12 GHz were also measured. The experiments were carried out on the HW1-Model Microwave Network Analyzer.

III. RESULTS AND DISCUSSION

Figure 1 shows the frequency dependence of the microwave-absorbing properties of $La_{1-x}Sr_xMnO_3$ ($x=0.4$, 0.5, 0.6, and 0.7) with the same coating thickness, $d=2.60$ mm, for the four different samples. Each curve shows an absorption loss peak near 10.5 GHz and the absorption peak values systematically decrease from 18.0 dB for $x=0.4$ to 4.8 dB for $x=0.7$. The microwave-absorbing frequency bandwidths defined as the frequency width in which the absorption is larger than 8 dB are about 3.0 GHz and 4.0 GHz for $x=0.4$ and 0.5 samples, respectively. Such wide absorption bandwidths and high absorption loss peaks indicate the attractive potential microwave applications. It should be noted that the absorption was measured for the samples mixed with wax by the ratio of 8:3 in weight. In fact, the actual absorbing losses must be much larger than the current values if the coatings are entirely made of those manganese oxide powders.

From Fig. 1, one can find that with the same coating thickness, the absorption loss peaks for $x=0.4$, 0.5, and 0.6 samples are much larger than that for $x=0.7$ sample. Maryland group[10,11] recently reported measurements of the microwave losses in microsize powders of several manganites. It was shown that for $La_{0.7}Sr_{0.3}MnO_3$ and $La_{0.7}Ba_{0.3}MnO_3$, the zero-field absorption exhibits a large increase as the temperature drops below a characteristic temperature $T^*(T^*<T_C)$ which is higher than room temperature. We know that both Curie temperatures for $La_{0.7}Sr_{0.3}MnO_3$ and $La_{0.7}Ba_{0.3}MnO_3$ are much higher than room temperature. Our samples with doping level $x=0.4$, 0.5, and 0.6 are in the ferromagnetic state and the other one ($x=0.7$) is in the paramagnetic state at room temperature.[6,7] So, the microwave loss for the $x=0.7$ sample is smaller than those for $x<0.7$ samples, which agrees with the Maryland group's results.[10,11] It is naturally considered that the losses are related to ferromagnetism: the formation of magnetic domains, and the resonance absorption of the moving magnetic domains and spin relaxation in the high frequency alternating electromagnetic fields.

In order to find the optimal matching frequency and coating thickness, we measured the absorption for the $x=0.4$ sample with different coating thicknesses (d varies from 2.0 to 2.6 mm), as shown in Fig. 2. It was found that the optimal microwave-absorbing properties are achieved when the thickness is 2.25 mm. The loss absorption peak value is about 25 dB around 9.7 GHz and the peak bandwidth is larger than 3.2 GHz. Simple calculation indicates that the incident microwave radiation is attenuated by more than 95%.

Usually, the microwave-absorbing properties of the ferrites are dominated by the magnetic and dielectric losses.[2] In order to know which factor dominates the absorption of the manganese oxides, one should examine the complex dielectric permittivity $\tilde{\varepsilon}(=\varepsilon'-i\varepsilon'')$ and magnetic permeability $\tilde{\mu}(=\mu'-i\mu'')$. Here, we measured the frequency dependence of the dielectric and magnetic absorption losses, tan $\delta_e(=\epsilon''/\epsilon')$ and tan $\delta_m(=\mu''/\mu')$, for $x=0.5$ coating of 2.6 mm in thickness. As shown in Fig. 3, tan δ_e and tan δ_m are large in the whole frequency range. It was found that tan δ_m

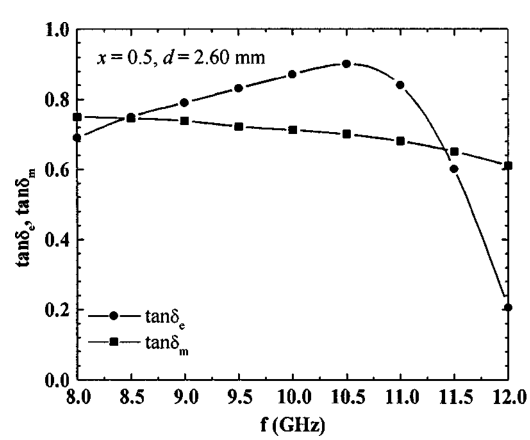

FIG. 3. The frequency dependence of the dielectric and magnetic absorption losses, tan δ_e and tan δ_m, for $x=0.5$ coating.

almost does not change with frequency (precisely speaking, it becomes slightly small with increasing frequency), while $\tan \delta_e$ shows a peak near 10.5 GHz corresponding to the absorption loss peak position for the sample. According to Ref. 13, the microwave loss can be divided into two parts: one is the resistive effect, and the other is spin-dependent loss due to the onset of ferromagnetism. It implies that the absorption can be ascribed to both dielectric and magnetic losses, namely, the absorption loss peak and frequency bandwidth are due to the dielectric and magnetic losses, respectively. As far as we know,[11–13] these features could not be well explained at present. Detailed analysis on the absorption properties is under way.

The dielectric loss for ferrites originates from the electron transfers between Fe^{2+} and Fe^{3+}, and the magnetic loss comes from the movements and spin relaxation of magnetic domains.[4] For a $La_{1-x}Sr_xMnO_3$ system, due to the divalent Sr ions introduced into the parent compound $LaMnO_3$, Mn^{3+} and Mn^{4+} mixed-valence ions appear in the system. According to the double exchange model,[14] the electron transfers between the Mn^{3+} and Mn^{4+} ions leading to the metallic conduction and ferromagnetism. This is very similar to that observed in ferrites. Naturally, the absorption for Sr-doped manganese oxides could be attributed to magnetic and dielectric losses. Further studies on the relationships among the absorbing properties, magnetization, resistivity, and magnetic domain structures of the compounds are in progress.

Many microwave-absorbing materials, like *Y*-type planar hexagonal ferrites, have been studied by mixing them with other materials, like carbonic fiber, for the purposes of deducing the weight of the absorbing materials and enhancing the absorption as well. In order to study the microwave-absorbing properties of the manganese oxides, we chose $x=0.7$ and $x=0.5$ samples since the former shows lower absorption while the latter shows higher absorption and wider absorption bandwidth, as shown in Fig. 1. For measuring microwave-absorbing properties, $x=0.7$ and $x=0.5$ manganese oxide powders were thoroughly mixed with the carbonic fiber and the *Y*-type planar hexagonal ferrite by the ratio of 1:1 in weight, respectively, to produce the square coatings. The thicknesses of the coatings are 2.6 mm for $x=0.7$ and 2.0 mm for $x=0.5$. From Fig. 4, one can see that the absorption loss for the $x=0.7$ sample is greatly enhanced almost by three times after the carbonic fiber is added, and the absorption peak moves to the higher frequency region. Similar phenomena also appear in the $x=0.5$ sample mixed with the *Y*-type planar hexagonal ferrite. For the carbonic fiber additive, the enhancement of absorption could be aroused by the dielectric loss of the carbonic fiber, while for the *Y*-type planar hexagonal ferrite additive, the increase of absorption may correspond to the natural resonant absorption of magnetic domains of the ferrite.[2] These features indicate that the manganese oxides can be mixed with other microwave absorbers to produce excellent composite microwave-absorbing materials.

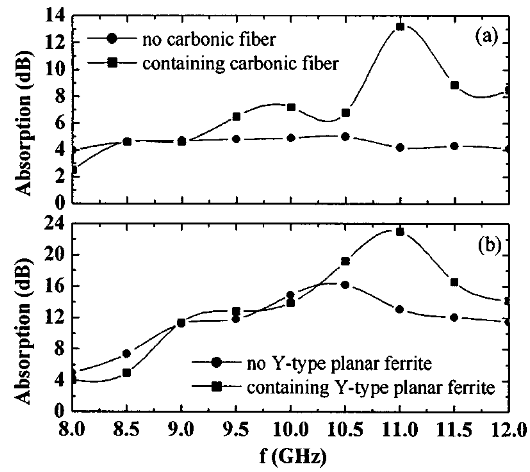

FIG. 4. The enhanced microwave-absorbing properties of manganese oxide powders for the $x=0.7$ (a) and $x=0.5$ (b) coatings after being mixed with the carbonic fiber and the *Y*-type planar hexagonal ferrite, respectively.

In summary, we reported the microwave-absorbing properties of $La_{1-x}Sr_xMnO_3$ ($x=0.4$, 0.5, 0.6, and 0.7) manganese oxides in the frequency range of 8–12 GHz at room temperature. Experimental results show that the optimal absorption peak is about 25 dB for $x=0.4$ coating. The investigation of complex magnetic permeability and dielectric permittivity indicates that the absorption can be ascribed to the magnetic and dielectric losses. The absorbing properties of the oxides have also been studied by mixing them with the carbonic fiber and *Y*-type planar hexagonal ferrite. These additives greatly enhance the absorption properties of the oxides.

ACKNOWLEDGMENT

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