

学位論文要旨

Electromagnetic Properties and Double
Negative Characteristics of Hybrid Granular
Composite Materials in the Microwave
Frequency Range

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Chapter 1: Introduction

The development of double negative materials (DNMs) with simultaneous negative value of permittivity and permeability has been the subject of considerable interest. It has been theoretically predicted and experimentally demonstrated that the left-handed (LH) or DNMs show extraordinary electromagnetic response such as the inverse Doppler Effect and negative index of refraction. These properties can lead to a diversity of optical or microwave applications such as modulators, super lenses, microwave couplers and antennas.

A number of studies have been carried out to realize the DNG composite materials using magnetic, dielectric or metallic alloys and compounds. The negative permeability (Mu Negative: MNG) can be obtained in some ferrites or magnetic metal composite materials by the magnetic resonance of ferromagnetic particles. To realize the negative permittivity (Epsilon Negative: ENG) the low frequency plasmonic state has to be achieved and this can be made by metallic substances.

Since the DNG properties in metamaterials originate from artificial structures rather than directly from natural materials. Therefore, it is also interesting to investigate the possibilities of realizing the DNG properties from the point of "real" random materials rather than periodic artificial structures. It has been proven that many "real" materials possess single negative permittivity or negative permeability. Hence, in the present thesis, hybrid granular composite materials including Cu metallic particles and ferromagnetic $\text{Fe}_{53}\text{Ni}_{47}$, $\text{Fe}_{50}\text{Co}_{50}$, Ni-Zn Ferrite, Mn-Zn Ferrite particles have been selected to provide negative permittivity ϵ and negative permeability μ , respectively.

Actually, few works exist on the realization of DNG characteristic using granular composite materials especially in frequency range above 1GHz. In

this thesis, the results of electric permittivity and magnetic permeability analysis in frequency range up to 20 GHz and simultaneous negative permittivity and negative permeability, DNG characteristic up to above 1 GHz is presented. Another significant point in the present study is that we are reporting for the first time the existence of DNG spectra of hybrid composite materials in the microwave range in the absence of the external magnetic field. The quasi-isotropic Negative Refractive Index (NRI) or Zero Index Material (ZIM) can be realized using the DNG granular composite materials.

The thesis is organized into five chapters starting from the current chapter one which intends to give a brief introduction of the historical development of LH materials and the motivation of this study, followed by the chapter two, which presents the review on metamaterials; chapter three, the experimental method is presented; in chapter four, the experimental results is presented and discussed. Finally, the chapter five presents the conclusion of the findings.

Chapter 2: Theory on metamaterials - Review

Metamaterials are artificial homogeneous structures with unusual electromagnetic properties not readily available in nature. These structures are constructed with two or more materials at a macroscopic level. The materials are used to control and manipulate the electromagnetic parameters allowing the production of any value of electric permittivity and magnetic permeability. In general, electromagnetic (EM) response in homogeneous materials is predominantly governed by the two parameters, permittivity ϵ and permeability μ . Both parameters are typically frequency-dependent complex quantities

In general, there is four possible sign combinations of the ϵ and μ pair. The first one, $\epsilon > 0$ and $\mu > 0$, (+,+) double positive (DPS) materials; the second

and third combinations correspond to the single negative (SNG) materials, (-, +), $\epsilon < 0$ and $\mu > 0$, ENG; (+,-), $\epsilon > 0$ and $\mu < 0$, MNG and the last combination (-,-), $\epsilon < 0$ and $\mu < 0$, DNG materials.

A LH material is an electromagnetic medium with simultaneous negative ϵ and μ . The DNG properties result in the propagation of electromagnetic waves exhibiting antiparallel phase and group velocity or LH waves. In a RH medium the wave vector number \vec{k} is positive and goes outward propagation from the source. In a LH, \vec{k} is negative and the wave goes inward propagation to the source.

The permeability and permittivity dispersion in a material can be described by the Lorentz model for dielectric permittivity and magnetic permeability and by the Drude model which is especial case of Lorentz model. In the Lorentz model the electrons are bound tightly to their nucleus and hence will not be free to move around and contribute to the materials conduction while in the Drude model, the electrons are free to move making in this way the electrical conduction. Since the composite materials are constituted of two or more materials; hence, they are inhomogeneous. Maxwell Garnett has developed a homogenization theory to approximate a complex electromagnetic medium, the effective medium approximation to describe the composite material as a whole. The Maxwell Garnet Approximation (MGA) formula gives the effective permittivity and effective permeability of this effective medium. Another mixing formula, the Coherent Model Approximation (CMA), is used to analyze the complex permeability. Granular composite materials contain various clusters and insolated particles. Doyles and Jacobs investigates the change of dielectric to metallic properties in the suspension of conducting spheres using the effective cluster model (ECM) based on the Clausius-Mossotti relation and evaluate the dielectric enhancement in the metal granular composite.

The DNG Characteristic in hybrid granular composite materials can be

attributed to the magnetic resonance (MR) originated by the combination of domain wall (DW) vibration and gyromagnetic spin rotation produced by the ferromagnetic particles and by the low frequency plasmonic state (LFP) produced by the metallic particles.

Chapter 3: Experimental Method

Based on "real" random materials, hybrid composite materials consisting of the combination of metallic Cu and magnetic FeNi, FeCo, NiZn Ferrite MnZn Ferrite particles were fabricated. The granular composite metamaterial samples were prepared by mixing ferromagnetic particles, metallic particles with Polyphenylene Sulfide (PPS) resin powder. The mixture was melting at 300⁰ C and pressing at pressure of 620 MPa in the cooling process down to the room temperature. Toroidal samples with inner diameter 3 mm and outer diameter of 7 mm were prepared which were used to collect the relative permittivity and permeability spectra data. For the electrical conductivity measurement, rectangular samples with 2 mm thickness were prepared. The composition of the selected hybrid granular composites is denoted by $(A_xB_{1-x})_yPPS_{1-y}$, where x is the volume fraction of A in the AB hybrid particle and y indicates the total volume fraction of embedded particles y = 0.85 in the case of Fe₅₀Co₅₀/Cu and Fe₅₃Ni₄₇/Cu and 0.80 for NiZn Ferrite/Cu and MnZnF/Cu granular composite materials. The relative complex permittivity ϵ_r and permeability μ_r of the composite materials were measured by the transmission/reflection method using a coaxial line cell and a network analyzer in the frequency range from 10 MHz to 20 GHz. The electrical conductivity of composite materials was measured by the two-terminal method using an impedance analyzer in the frequency range from 1 kHz to 40 MHz.

Chapter 4: Results and Discussion

The materials developed was analyzed by measuring the electrical conductivity in frequency range from 1 kHz to 40 MHz and complex permittivity and permeability spectra in the frequency range from 10 MHz to 20 GHz using a network analyzer and a transmission line approach.

The $\text{Fe}_{53}\text{Ni}_{47}/\text{Cu}$ and $\text{Fe}_{50}\text{Co}_{50}/\text{Cu}$ composites are considered to have a double metallic system since the ferromagnetic particles $\text{Fe}_{53}\text{Ni}_{47}$ and $\text{Fe}_{50}\text{Co}_{50}$ combined with Cu particles contributes to the electrical properties. Thus, are treated as a single particle system in the host PPS resin. While in the case of NiZn ferrite/Cu and MnZn Ferrite/Cu, the ferrite particles have low conductivity, the electrical properties of the composites are determined mainly by the Cu particles content against the total volume. In general, the electrical percolation effect is determined mainly by the volume fraction of Cu metallic particles. Increasing the Cu particles content, the conductivity is enhanced, LFP state is achieved and the negative permittivity takes place. The negative permeability is realized by the magnetic resonance of $\text{Fe}_{53}\text{Ni}_{47}$, $\text{Fe}_{50}\text{Co}_{50}$ ferromagnetic and NiZn and MnZn Ferrite particles. The frequency dispersion of permittivity was evaluated by the numerical fitting of the measuring ϵ_r' using Drude model for metals and Lorentz model for dielectrics. The fitting results showed a fairly good agreement between numerical and experimental data in the ϵ_r' spectrum but a relatively large discrepancy exists between calculated and measured data in the imaginary parts ϵ_r'' . The origin of the discrepancy might be attributed to the other dielectric energy dissipation process in the metal composite structure. The variation of the low frequency permittivity of NiZn Ferrite/Cu composites with particle content was evaluated by the ECM. A relatively large discrepancy between experimental and numerical data was observed. However, the qualitative variation of dielectric constant with particle content can be described by the ECM model for the NiZn

Ferrite/Cu composite materials.

Since the ENG spectrum was realized by the low frequency plasma oscillation of conduction electrons in the percolated Cu cluster chain, ENG was obtained in the wide frequency range below f_0 and the negative ε_r' value become large in the low frequency range. Meanwhile, the MNG spectrum was obtained by the magnetic resonance in the ferromagnetic and ferrite particles, the negative μ_r' band is narrow and its value is relatively small.

The DNG characteristic was realized from the sub microwave range to about 1.6 GHz and 2 GHz in the $\text{Fe}_{53}\text{Ni}_{47}/\text{Cu}$ and NiZn Ferrite/Cu hybrid composite materials respectively. While the DNG characteristic was realized in the sub microwave and X-band range in the $\text{Fe}_{50}\text{Co}_{50}/\text{Cu}$ and MnZn Ferrite/Cu hybrid composite materials in frequency range up to 10 GHz. The DNG band can be controlled by changing the Cu and ferromagnetic particles content ratio in the granular composite structure.

Chapter 5: Conclusions

DNG characteristic was realized in the sub microwave and X-band range in frequency range up to 10 GHz in the absence of external magnetic field.

The composite materials in this study have relatively high conductive loss (imaginary part of permittivity shows a large value) in the percolation-induced LFP state in around the DNG band. Electromagnetic wave energy can be easily dissipated in these materials. This indicates that the transmission of the electromagnetic wave through the composite at the DNG band can become small. Hence, the negative permittivity characteristic can be utilized for the construction of meta-surface with the frequency selective transmission property. The electromagnetic wave shieldings or absorbing applications can be considered to use these DNG composites as well as the negative refraction materials. The quasi-isotropic Negative Refractive Index or Zero Index

Material can be realized using the DNG granular composite materials

The calculated NRI spectrum for the $x = 0.24$ in $(\text{Cu}_x\text{FeNi}_{1-x})_{0.85}\text{PPS}_{0.15}$, $\text{Fe}_{53}\text{Ni}_{47}/\text{Cu}$ composite as function of frequency was examined and the NRI was obtained from 200 MHz to 1.8 GHz.

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