Influence of Bi and Sb cations on the electric and magnetic properties of Mg doped copper ferrites

Thesis
Submitted for the degree of Ph.D.
In Solid State Physics

By **Ali Ahmed Mohamed Azab**

To
Physics Department
Faculty of Science
Cairo University

2010

تأثير كاتيونات البزموث و الأنتيمون على الخواص الكهربية و المغناطيسية لفريت النحاس المطعم بالماغنسيوم

رسالة مقدمه لقسم الفيزياء – كلية العلوم – جامعة القاهرة

> لنيل درجة الدكتوراة فيزيقا الجوامد

على أحمد محمد عزب

Abstract

The investigated samples were prepared by two techniques the standard and wet methods (sol-gel, citrate and co precipitation). Three groups were prepared by the standard ceramic technique with the formula.

- 1- The first group has the formula $Cu_{1-x} Mg_x Fe_2 O_4$ where $0 \le x \le 0.6$
- 2- The second group has the formula Cu $_{0.7}$ Mg $_{0.3}$ Sb $_y$ Fe $_{2\text{-y}}$ O $_4$ where $~0.05 \leq ~y \leq 0.25$
- 3- The third group has the formula $Cu_{0.7}$ Mg $_{0.3}$ Bi $_z$ Fe $_{2-z}$ O $_4$ where $0.05 \le z \le 0.25$ The sample with x=0.3 ($Cu_{0.7}$ Mg $_{0.3}$ Fe $_2$ O $_4$) was prepared by the wet methods under different conditions.

X-ray diffraction patterns show that, the first group showed cubic spinel structure with small intensities of secondary phase at $0 \le x \le 0.2$ after which no secondary phases were exist, in the second group, secondary phases appears at y=0.15, 0.25. While in the third group single phase cubic spinel structure was obtained. The sample with x=0.3 was prepared by co precipitation method at different PH values (9-13.5) and it is sintered at different temperatures (200-950 °C) for 3 hrs. Also the sample with x=0.3 was prepared by citrate and sol-gel methods. The lattice parameter and X-ray density for the above mentioned samples were calculated and reported.

The IR spectra for the prepared samples, were carried out in the rang of 200-700 cm⁻¹ in order to confirm the formation of the samples in the proper form. Four bands indicating the spinel structure were obtained, two of them v_1 , v_2 are of high frequency and the other two v_3 , v_4 are of low frequency.

The transmission electron microscope (TEM) was performed to explore the particle shape and size, micrographs reveals different shapes, homogeneities and particle size. Sol-Gel method gave the smallest particle size (8.7 nm), while the ceramic method gave the largest particle size (127.8 nm).

The electrical conductivity was measured at different temperatures as a function of frequency (10 kHz–5MHz). The AC conductivity increases with increasing temperature giving three regions; the first one is stable region (flat region) while the other two are with different slopes indicating the different conduction

mechanisms. The general trend was a decrease in the conductivity with increasing the concentration of the cations Mg^{2+} , Sb^{3+} and Bi^{3+} .

The real part (ϵ') and the imaginary part (ϵ'') of dielectric constant were studied as a function of temperature and frequency (100 kHz – 5MHz). The variation of the dielectric constants depends mainly on the valence exchange between the different metal ions either of one element or two different elements. The parameters ϵ' and ϵ'' showed a decrease with increasing of Mg^{2+} , Sb^{3+} and Bi^{3+} concentrations and an increase with increasing temperature.

Seebeck coefficient measurements were carried out with temperature to investigate the type of charge carriers. Generally, we have found that the small polarons as well as the electrons contribute to the conduction process. The use of the small polaron hopping interprets the positive values obtained for Seebeck coefficient of most samples.

The molar magnetic susceptibility (χ_M) for the samples was studied at different temperatures as a function of the magnetic field intensities. The magnetic parameters such as Curie temperature (T_C) and the effective magnetic moment (μ_{eff}) were calculated from the experimental data and reported. The data showed that, all parameters were decreased with increasing Mg^{2+} , Sb^{3+} and Bi^{3+} concentrations. This is due to the replacement of the paramagnetic and ferromagnetic cations with diamagnetic ions, on octahedral site which decreases the A-B exchange interaction.

The magnetic parameters obtained from hystersis for the sample prepared by different methods (ceramic, sol- gel, coprecipitation and citrate) show that, they changed depending on the preparation methods. High coercivity was obtained for the nanorods shaped sample (citrate method) 125.6 Oe. Highest saturation and remanant magnetization (9.87 emu/g) was achieved for the sample prepared by the standard ceramic technique. Finally one can control the magnetic characterizations by using the suitable method of preparation.



جامعــة القـــاهرة

استمارة معلومات الرسائل التي تمت مناقشتها



١ ـ الدرجة العلمية: دكتوراه

٢ - بيانات الرسالة:

القسم: الفيزياء

الكلية: العلوم

عنوان الرسالة باللغة العربية:

تأثير كاتيونات البزموث و الأنتيمون على الخواص الكهربية و المغناطيسية لفريت النحاس المطعم بالماغنسيوم

عنوان الرسالة باللغة الأجنبية: Influence of Bi and Sb cations on the electric and magnetic properties of Mg doped copper ferrites

التخصص الدقيق: فيزياء جوامد تاريخ المناقشة:

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الجنسية: مصري

۳_ بيانات الطالب: الاسم: على أحمد محمد عزب

جهة العمل: المركز القومي للبحوث رقم الفاكس:

٤ ـ المشرفون على الرسالة:

| الجامعة | الكلية | القسم | الاسم |
|------------|-------------|----------------|--------------------------------|
| القاهرة | العلوم | فيزياء | ۱ - ا.د. محمد على أحمد |
| القاهرة | العلوم | فيزياء | ٢- د علاء الدين عبد اللطيف |
| ِمي للبحوث | المركز القو | فيزيقا الجوامد | ٣- أ. د حسن حسن عفيفي |
| رمى للبحوث | المركز القو | فيزيقا الجوامد | ٤- أبد إيناس أحمد كمال الزواوي |



جامعــة القـــاهرة

٥ ـ مستخلص الرسالة:

٥-١ باللغة العربية:

الكلمات الدالة: فيريتات - نانو - نحاس - ماغنسيوم- بزموث - أنتيمون- نفاذيه مغناطيسيه

تم تحضير ثلاث مجموعات من المركبات بطريقة السير اميك ذات الصيغه

 $0 \leq x \leq 0.6$ حيث $Cu_{1-x} \ Mg_x \ Fe_2 \ O_4$ حيث الأولى لها الصيغة 1

 $0.05 \leq y \leq 0.25$ حيث $Cu_{0.7}$ Mg $_{0.3}$ Sb $_y$ Fe $_{2-y}$ O $_4$ حيث $Cu_{0.7}$ Hg حيث $Cu_{0.7}$ المجموعة الثانية لها الصيغة

 $0.05 \le z \le 0.25$ حيث $Cu_{0.7} \, \mathrm{Mg}_{0.3} \, \mathrm{Bi}_z \, \mathrm{Fe}_{2-z} \, \mathrm{O}_4$ حيث -7

العينة التي لها الصيغة (${\rm Cu}_{0.7}\ {
m Mg}_{0.3}\ {
m Fe}_2\ {
m O}_4$) تم تحضير ها بطرق الكيمائية و تحت ظروف تحضير مختلفة للحصول على عينات نانو متريه.

تم استخدام حيود الأشعة السينية و تحت الحمراء في المدى 1 200cm الى 2 700 km التأكد من تكون العينات في الصورة Spinel . تم أخذ صور للعينات بالميكر سكوب الآلكتروني النافذ لمعرفة حجم و شكل البلورات وقد وجد أن طريقة sol 30l-gel أعطت أصغر حجم بلوري (sol 17۷ نانومتر) و طريقة السيراميك القياسية أعطت أكبر حجم بلوري (sol 17۷ نانومتر).

تم قياس التوصلية الكهربية المترددة والجزء الحقيقى والجزء التخيلي لثابت العزل الكهربي

كدالة في درجة الحرارة و قيم مختلفة للتردد (١٠ كيلو هرتز - ٥ ميجا هرتز).

تم قياس القابلية المغناطيسية و حساب الثوابت المغناطيسية من منحنى عروة التخلف المغناطيسي للعينات المحضرة بطرق مختلفة و قد وجد من النتائج أن قيم الثوابت المغناطيسية تعتمد على طرق التحضير و قد أعطت طريقة Citrate شكل قضبان بلورية نانومترية أعلى قيمة للمجال القسرى (٦.٥٦ أورستد).



جامعة القاهرة

٥-٢ باللغة الأجنبية: (Abstract)

Key Words: Ferrite, nanoferrite, copper, magnesium, bismuth, antimony, magnetic susceptlity, thermo eclectic power, conduction mechanism.

Three groups of investigated samples were prepared by the standard ceramic technique with the formula.

- 1- The first group has the formula Cu_{1-x} Mg _x Fe₂ O₄ where $0 \le x \le 0.6$
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The sample with x=0.3 (Cu0.7 Mg 0.3 Fe₂ O₄) was prepared by the wet methods under different conditions to get nano-particle size. X-ray diffraction patterns and IR spectra for the prepared samples, were carried out in order to confirm the formation of the samples in the spinel structure. The transmission electron microscope (TEM) was performed to explore the particle shape and size. Sol-Gel method gave the smallest particle size (8.7 nm), while the ceramic method gave the largest particle size (127.8 nm). The ac electrical conductivity, the real part (ε') and the imaginary part (ε'') of dielectric constants were studied as a function of temperature and frequency (100 kHz – 5MHz). The molar magnetic susceptibility ($\chi_{\rm M}$) was studied at different temperatures as a function of the magnetic field intensities. The magnetic parameters obtained from hysteresis for the sample prepared by different methods (ceramic, sol- gel, coprecipitation and citrate) show that, they changed depending on the preparation methods. High coercivity was obtained for the nanorods shaped sample (citrate method) 125.6 Oe.



جامعــة القـــاهرة

٦- أهم النتائج التي تم التوصل إليها:

- 1-7 من الخواص الكهربيه و جد ان العينات تعطى مقاومة كهربيه عاليه و من الخواص المغناطيسيه وجد القيم المنخفظة H_C تجعل من المادة ملائمة لعمل قلب المحولات الكهربية ذات القدرات العاليه.
 - L-C من الخواص الكهربيه و العزليه تجعل من الماده مناسبة L-C في الدوائر الكهربيه
 - 7-7 الماده لها قدرة امتصاص للموجات الكهرومغناطيسيه تجعلها ملائمة في عمل تغطيه لكابلات البيانات لمنع التداخل بين الاشارات.
 - ٦-٤ المادة لها خواص للتطبيق كمستشعر للرطوبة.



جامعة القساهرة

٧- ما هي الجهات التي يمكن أن تستفيد من هذا البحث: الصناعات الكهربيه و الالكترونيه (المحولات الكهربيه – الدوائر الالكترونيه)

- الاتصالات (كمبيوتر - وكابلات نقل البيانات)

٨ - هل توجد علاقة بإحدى هذه الجهات: لا

٩- هل توافق على التعاون مع جهات مستفيدة من خلال الجامعة: نعم

- (أ) لتطبيق البحث.
- (ب) لاستكمال البحث.
- ١٠ ـ هل تم نشر بحوث مستخرجة من الرسالة في مجلات أو مؤتمرات علمية: لا
 - ١١- هل تم سبق التقدم لتسجيل براءات اختراع: لا
- ١٢- هل توافق على إعطاء البيانات المذكورة في هذه الاستمارة لجهات أخرى: نعم

توقيع الطالب/ توقيع المشرفين: على أحمد محمد عزب

وكيل الكلية للدراسات العليا والبحوث:

التاريخ:

١١٧ م العامة للدر اسات العليا



| لإداره العامة للدر اسات العلي والبحوث | 1908 2008 (1) 2008 | جامعــة القــــاهرة ــــــــــــــــــــــــــــــــــــ |
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Cairo University Faculty of Science

Directory of Postgraduate Theses Ph.D.

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Registration Date: Awarding Date: Supervisors:

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 Bahauddin Zakariya University, Pakistan

Title of Thesis: Influence of Bi and Sb cations on the electric and magnetic properties of Mg doped copper ferrites

Key words: Ferrite, nanoferrite, copper, magnesium, bismuth, antimony, magnetic susceptlity, thermo eclectic power, conduction mechanism

Summery:

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Approval Sheet

Influence of Bi and Sb cations on the electric and magnetic properties of Mg doped copper ferrites

Name of the candidate

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Influence of Bi and Sb cations on the electric and magnetic properties of Mg doped copper ferrites

Thesis

Submitted for the degree of **Ph. D.** in Solid State Physics

By Ali Ahmed Mohamed Azab

To

Physics Department Faculty of Science Cairo University

2008

رسالة مقدمة لقسم الفيزياء - كلية العلوم - جامعة القاهرة

لنيل درجة الدكتوراه

على أحمد محمد عزب