May 5, 1964 G. R. HARRISON ETAL 3,132,105

TEMPERATURE COMPENSATED YTTRIUM GADOLINIUM IRON GARNETS

Filed March 14, 1962 2 Sheets-Sheet 1

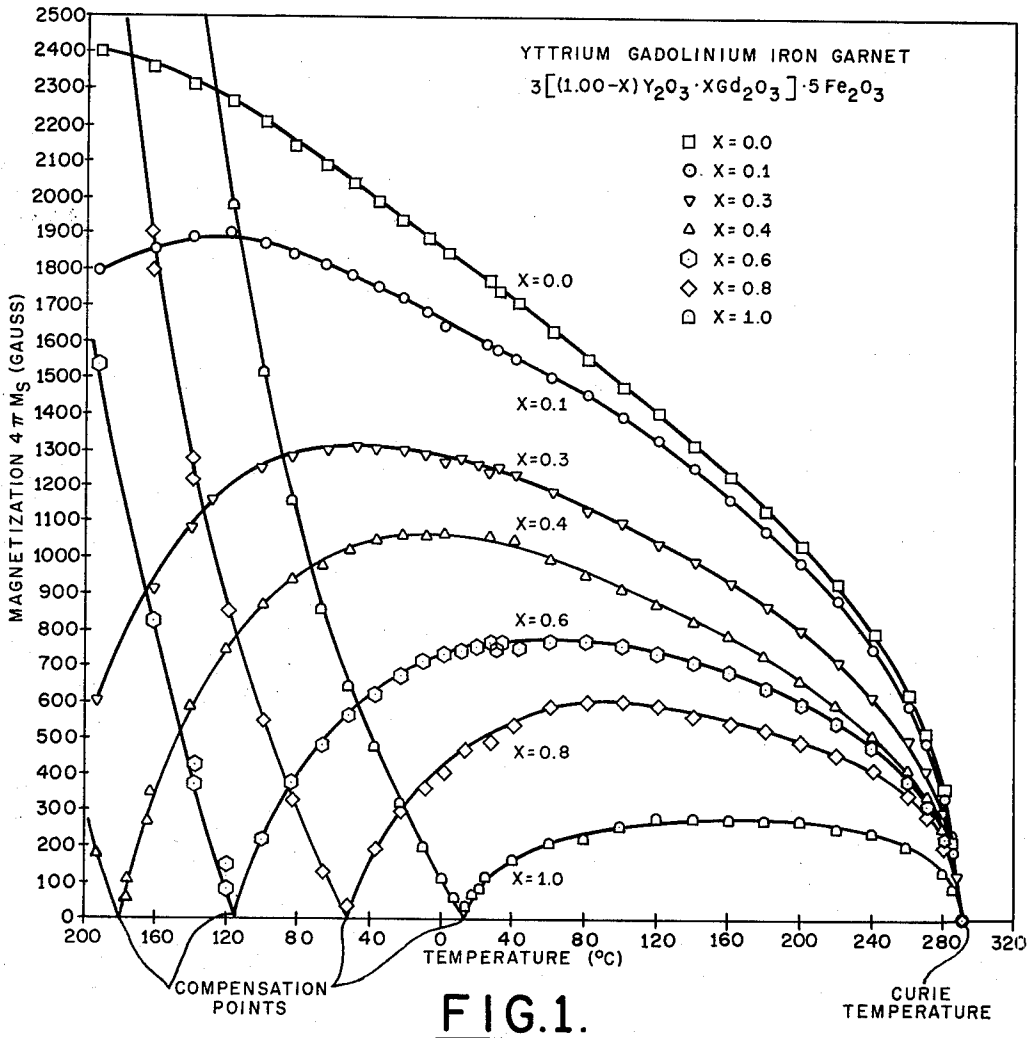


FIG.1.

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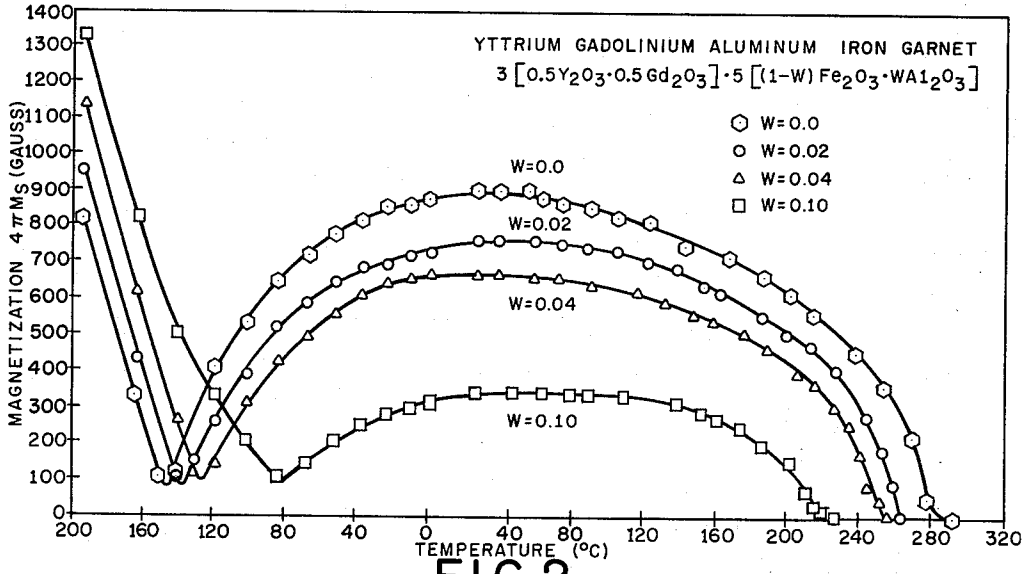


FIG.2.

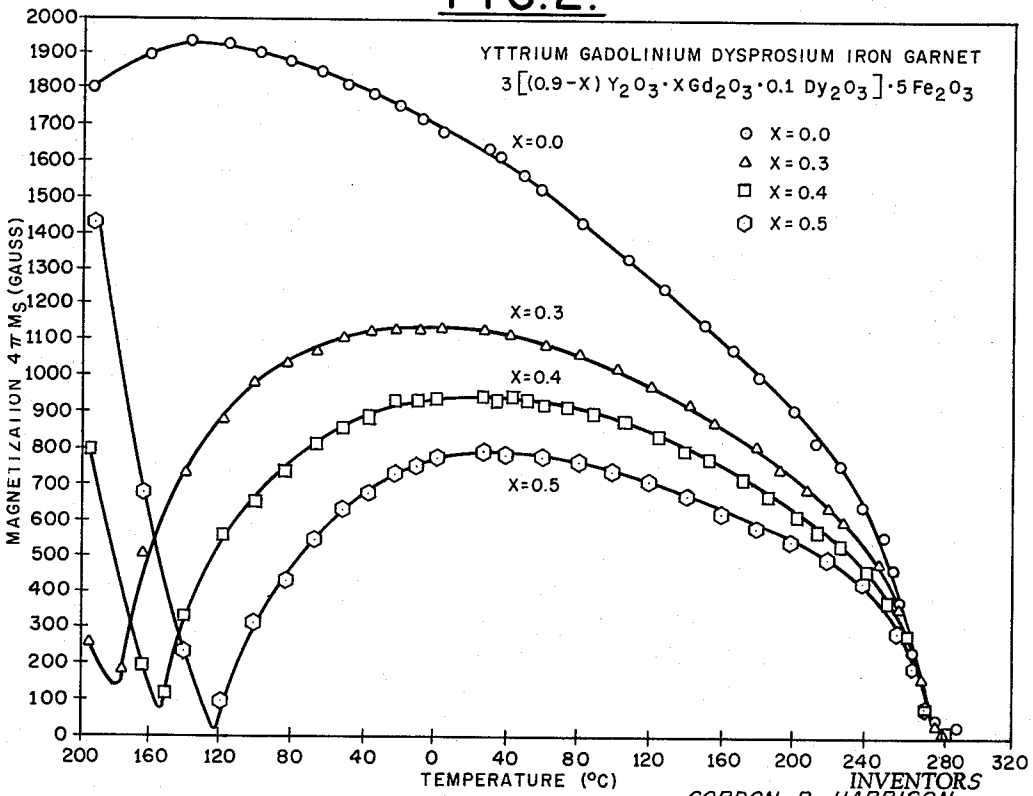


FIG.3

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3,132,105

# TEMPERATURE COMPENSATED YTTRIUM GADOLINIUM IRON GARNETS

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7 Claims. (Cl. 252—62.5)

The present invention generally relates to microwave garnet materials and, more particularly, to yttrium gadolinium iron garnets of improved temperature stability and high power handling capability.

Ferrimagnetic garnet materials have stimulated much interest since their discovery, especially with respect to their applicability to present microwave devices. This interest has led to intensive studies of the crystallographic, magnetic and other fundamental properties of these materials. Less attention has been given heretofore to properties which are primarily significant from a microwave engineering point of view. Of the ferrimagnetic garnet compositions developed to date, yttrium iron garnet appears to be the most promising for microwave applications. It has been found, however, that pure yttrium iron garnet does not adequately satisfy certain microwave desiderata including temperature stability and power handling capability. More particularly, there is a need for improving yttrium iron garnet material so as to render its saturation magnetization characteristic substantially independent of temperature over a wide range of temperature values including room temperature and to extend its high power handling capabilities. It is important, of course, that the improvements desired be achieved without undue sacrifice respecting other pertient microwave properties such as line-width, $g$-factor and Curie temperature.

It is the principal object of the present invention to provide solid solutions of ferrimagnetic garnet materials having improved temperature stability.

Another object is to provide solid solutions of ferrimagnetic garnet materials having increased microwave power handling capability.

A further object is to provide ferrimagnetic garnet materials characterized by impoved temperature stability and increased power handling capability.

An additional object is to provide a mixed yttrium gadolinium iron garnet having a relatively low saturation magnetization value which remains substantially constant over an extended range of temperature values.

These and other objects, as will appear from the reading of the following specifications, are accomplished in a first aspect of the invention by mixing yttrium, gadolinium, iron oxide, and one of the metals aluminum and gallium in special proportions to produce a composite garnet material. When the proportions of the constituent materials are established in accordance with the relationships discussed herein, the saturation magnetization characteristics of the first composite garnet material is stabilized at values as low as about 300 gauss over a temperature range from about —30 to about 140° C.

In another aspect, the invention provides for the mixture of yttrium, gadolinium, dysprosium and iron oxide in special proportions, to provide a composite garnet material characterized by a saturation magnetization of the order of 900 gauss which remains substantially constant over the temperature range from about —20° to about 100° C. Additionally, the power handling capability of the composite garnet material is increased by two orders of magnitude as compared to pure yttrium iron garnet.

In a third aspect, the invention provides for a solid solution of yttrium, gadolinium, dysprosium, iron oxide and one of the metals aluminum and gallium in special



proportions to yield a composite garnet material possessing a wide range of saturation magnetization values, each of which is temperature stabilized in addition to having improved high power handling capability.

For more complete understanding of the present invention, reference should be made to the following specification and to the figures of which:

FIG. 1 is a plot of the variation of saturation magnetization with respect to temperature for yttrium gadolinium iron garnet;

FIG. 2 is a plot of the variation of saturation magnetization with respect to temperature for yttrium gadolinium aluminum iron garnet and;

FIG. 3 is a plot of the variation of saturation magnetization with respect to temperature for yttrium gadolinium dysprosium iron garnet.

The material geometries employed in most microwave devices are such that the frequency of ferrimagnetic resonance is not only dependent on the magnetic field but also on the saturation magnetization of the material. Consequently, it is desirable in the design of and development of microwave ferrimagnetic devices which are to operate over large temperature ranges that the magnetization of the material utilized be stabilized with respect to temperature. As can be seen from inspection of FIG. 1, a certain degree of magnetization stabilization may be obtained by the appropriate proportioning of the yttrium and gadolinium constituents of yttrium gadolinium iron garnet.

The magnetization vs. temperature characteristics depicted in FIG. 1 are those of polycrystalline garnets of the form $3[(1.00-x)Y_2O_3 \cdot xGd_2O_3] \cdot 5Fe_2O_3$. The data is plotted over a temperature range from about —195° C. to the Curie temperature of the compositions. The curves differ from each other due to a change in the proportioning of the yttrium and gadolinium constituent materials. It will be noted that the magnetization values above about 15° C. decrease as the gadolinium content ($x$) increases and that the compensation points (when present) appear at relatively higher temperatures as $x$ increases. It should be particularly noted that when $x=0.6$, the polycrystalline garnet materials exhibit a relatively stable saturation magnetization over a large temperature range. Other microwave properties of this particular composition have been found to be as follows:

Linewidth $\Delta H$ (25° C., X-band)=200 oersteds;
$g_{eff}$-factor (25° C., X-band)=2.08;
Dielectric loss tangent (1 gc.)=0.004; and
Dielectric constant (1 gc.)=13.

Although a substantial degree of temperature stabilization of saturation magnetization is achieved when the yttrium and gadolinium constituent materials are in the proportion of 2:3, little control over the value of the stabilized magnetization can be effected. In accordance with the present invention, however, the stabilized value of the constant magnetization may be controlled over extended ranges by the introduction of certain amounts of one or more of aluminum, gallium and dysprosium in the yttrium gadolinium materials whose characteristics are plotted in FIG. 1. In particular, the value of the temperature stabilized magnetization parameter may be controllably decreased from the 750 gauss value of FIG. 1. Such a decrease is especially significant in certain microwave applications of the composite garnet material including microwave phase shifter applications.

The magnetization vs. temperature characteristics plotted in FIG. 2 are those of a typical solid solution of yttrium gadolinium garnet material wherein relatively small amounts of aluminum are substituted for the iron in accordance with the expression

$$3[0.5Y_2O_3 \cdot 0.5Gd_2O_3] \cdot 5[(1-w)Fe_2O_3 \cdot wAl_2O_3]$$

For small substitutions, the aluminum locates predominantly on the 24*d* site in the garnet structure. As can be seen in FIG. 2, this reduces the resultant magnetization at 25° C., reduces the Curie temperature, and positions the compensation point to higher temperatures as the aluminum content ($w$) increases. The result is a series of polycrystalline garnet materials exhibiting controllable, temperature stable magnetizations from about 900 gauss downward. Other microwave properties of these compositions have been found to be as follows:

Linewidth $\Delta H$ (25° C., X-band)=165 to 300 oersteds;
$g_{eff}$-Factor (25° C., X-band)=2.04 to 2.10; and
Dielectric loss tangent (1 gc.)=0.004.

True compensation points were not observed for these compositions presumably due to a slight nonuniformity in composition from grain to grain. More generally, it has been found that controllable values of magnetization may be stabilized with respect to temperature in accordance with the expression

$$3[(1.00-x)Y_2O_3 \cdot xGd_2O_3] \cdot 5[(1.00-y)Fe_2O_3 \cdot yAl_2O_3]$$

where the values of $x$ are in the range from about .4 to about .7 and the values of $y$ are in the range from about .02 to about .15. Similar results may be achieved by the substitution of gallium for aluminum in accordance with the foregoing expressions.

A series of garnet materials having greater high power thresholds (superior power handling capability) can be obtained by the introduction of certain amounts of dysprosium. In accordance with the present invention, such garnet materials may be defined by the expression

$$3[(1.00-x-y)Y_2O_3 \cdot xGd_2O_3 \cdot yDy_2O_3] \cdot 5Fe_2O_3$$

where the values of $x$ are in the range from about .3 to about .50 and the values of $y$ are in the range from about .01 to about .15. The magnetic properties of a number of typical materials defined by the foregoing expression are slotted in FIG. 3. By inspection of FIG. 3, it can be seen that controllable values of magnetization are temperature stabilized for values of $x$ between about 0.3 and about 0.5. Other microwave properties of these materials have been found to be:

Linewidth $\Delta H$ (25° C., X-band)=450 to 700 oersteds;
$g_{eff}$-Factor (25° C., X-band)=1.80 to 1.76; and
Dielectric loss tangent (1 gc.)=0.004.

Aluminum or gallium may also be added for the further control of the stabilized magnetization levels as discussed in connection with FIG. 2. The resulting composite garnet material may be defined by the expression

$$3[(1.00-x-y)Y_2O_3 \cdot xGd_2O_3 \cdot yDy_2O_3] \cdot 5[(1.00-w)Fe_2O_3 \cdot w(TM)_2O_3]$$

X and $y$ are any decimal numbers between the values of about 0.3 and about 0.50 and between the values of about 0.01 and about 0.15, inclusive, respectively. $w$ is any decimal number between the values of about .02 and about 0.15 inclusive and (TM) represent the metals of aluminum (Al) or gallium (Ga).

The polycrystalline garnet materials of the present invention may be prepared in accordance with conventional sintering techniques. One suitable technique is described in the Microwave Journal, volume 4, number 6, page 53 (1961).

While the invention has been described in its preferred embodiments, it is understood that the words which have been used are words of description rather than of limitation and the changes within the purview of the appended claims may be made without departing from the true scope and spirit of the invention in its broader aspects.

What is claimed is:

1. The composition of matter which is the solid solution of yttrium iron garnet and gadolinium iron garnet wherein one element selected from the metals group consisting of aluminum and gallium is substituted in place of part of the iron in accordance with the expression

$$3[(1.00-x)Y_2O_3 \cdot xGd_2O_3] \cdot 5[(1.00-y)Fe_2O_3 \cdot y(ME)_2O_3]$$

where $x$ and $y$ are any decimal numbers within the range of values from about .4 to about .7 and from about .02 to about .15 inclusive, respectively, and ME is the element selected from said group.

2. The composition of matter which is the solid solution of yttrium iron garnet and gadolinium iron garnet wherein aluminum is substituted in place of part of the iron in accordance with the expression

$$3[(1.00-x)Y_2O_3 \cdot xGd_2O_3] \cdot 5[(1.00-y)Fe_2O_3 \cdot yAl_2O_3]$$

where x and y are any decimal numbers within the range of values from about .4 to about .7 and from about .02 to about .15 inclusive, respectively.

3. The composition of matter which is the solid solution of yttrium iron garnet and gadolinium iron garnet wherein aluminum is substituted in place of part of the iron in accordance with the expression

$$3[0.5Y_2O_3 \cdot 0.5Gd_2O_3] \cdot 5[(1-w)Fe_2O_3 \cdot wAl_2O_3]$$

where $w$ is any decimal number within the range of values from about .02 to about .15 inclusive.

4. The composition of matter which is the solid solution of yttrium iron garnet, gadolinium iron garnet and dysprosium iron garnet in accordance with the expression

$$3[(1.00-x-y)Y_2O_3 \cdot xGd_2O_3 \cdot yDy_2O_3] \cdot 5Fe_2O_3$$

where $x$ and $y$ are any decimal numbers between the values of about .3 and about .50 and between the values of about .01 and about .15 inclusive, respectively.

5. The composition of matter which is the solid solution of yttrium iron garnet, gadolinium iron garnet and dysprosium iron garnet in accordance with the expression

$$3[(0.9-x)Y_2O_3 \cdot xGd_2O_3 \cdot O \cdot 1Dy_2O_3] \cdot 5Fe_2O_3$$

where $x$ is any decimal number between the values of about .3 and about .5 inclusive.

6. The composition of matter which is the solid solution of yttrium iron garnet, gadolinium iron garnet and dysprosium iron garnet wherein one element selected from the metals group consisting of aluminum and gallium is substituted in place of part of the iron in accordance with the expression

$$3[(1.00-x-y)Y_2O_3 \cdot xGd_2O_3 \cdot y(Dy)_2O_3)] \cdot 5[(1.00-w)Fe_2O_3 \cdot w(ME)_2O_3]$$

where $x$ and $y$ are any decimal number between the values of about .3 and about .50 and between the values of about .01 and about .15 inclusive, respectively, where $w$ is any decimal number between the values of about .02 and about .15, inclusive, and ME is the element selected from said group.

7. The composition of matter which is the solid solution of yttrium iron garnet, gadolinium iron garnet and dysprosium iron garnet wherein aluminum is substituted in place of part of the iron in accordance with the expression

$$3[(1.00-x-y)Y_2O_3 \cdot xGd_2O_3 \cdot yDy_2O_3] \cdot 5[(1.00-w)Fe_2O_3 \cdot wAl_2O_3]$$

where $x$ and $y$ are any decimal numbers between the values of about .3 and about .50 and between the values of about .01 and about .15 inclusive, respectively, and where $w$ is any decimal number between the values of about .02 and about .15, inclusive.

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