# Effect of Nd doping on Dielectric properties and Electrical Conduction in Gadolinium Gallium Garnet (GGG) single crystal

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**Abstract**—The dielectric properties (Dielectric constant 'ε' and Dielectric loss 'Tanδ') of a pure and Nd doped Gadolinium Gallium Garnet ($Gd_3Ga_5O_{12}$ and $Gd_3Ga_5O_{12}:Nd^{3+}$) single crystals were measured in the frequency range from 1 kHz to 1 MHz and in the temperature range from room temperature to 700°C. Electrical conductivity (σ) was calculated using the data on ε and tanδ. DC conductivity was also measured in the same temperature range. The electrical conductivity and the effect of Nd doping in these garnets is discussed in light of existing data.

**Index Terms**—Garnet, dielectric constant, dielectric loss, ac and dc conductivity

## 1 INTRODUCTION

Synthetic Rare Earth Garnets have a general chemical formula '$A_3B_2C_3O_{12}$'. 'A' represents a Rare earth element such as $Gd^{3+}$, $Y^{3+}$, $Sc^{3+}$ etc. while 'B' and 'C' represents a Group III element such as $Ga^{3+}$, $Al^{3+}$, or $Fe^{3+}$ and the corresponding garnet is generally named as Gallium, Aluminium or Iron Garnet respectively.

Rare earth garnets belong to a space group '$Ia\bar{3}d$' in a Cubic structure with 8 molecules per unit cell. i.e., each unit cell contains $(A_3B_2C_3O_{12})_8$ with 160 atoms, with a lattice parameter around 12Å. Within the cube there are 24 dodecahedral 'c' sites occupied by Rare Earth ions such as $Gd^{3+}$, $Y^{3+}$, $Sc^{3+}$ etc. surrounded by 8 Oxygen ions, 16 Octahedral 'a' sites occupied by $Fe^{3+}$, $Sm^{3+}$ etc. surrounded by 6 Oxygen ions and 24 Tetrahedral 'd' sites occupied by $Al^{3+}$, $Ga^{3+}$ etc. surrounded by 4 Oxygen ions. Within the frame work of cubic structure under certain conditions it provides for anisotropy leading to interesting properties.

## 2 EXPERIMENTAL DETAILS

Highly polished single crystals of pure and Nd doped Gadolinium Gallium Garnets obtained from Litton Airtron, USA, were used in the dielectric investigations. Each crystal is in circular shape having dimensions of 1cm in diameter and 0.2cm in thickness. The crystal faces are cut parallel to (111) direction.

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Pure GGG is in white transparent colour while the Nd doped GGG is in pale pink colour. The crystal faces are coated with air-drying silver paint for better electrical contact with the electrodes.

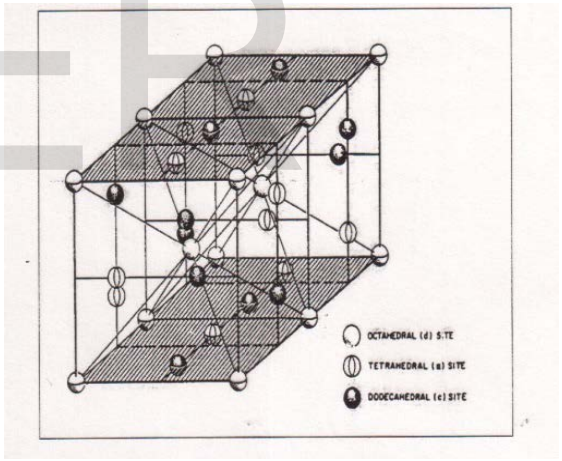


Fig.1 Array of cations in two octants of a garnet cell. [1]

## 3 RESULTS

The crystals were annealed in air at 700° for several hours before taking the measurements. Initially the dielectric measurements were done at room temperature. The variation of dielectric constant (ε) with frequency at room temperature for pure and Nd doped GGG are shown in figures 2 and 3.

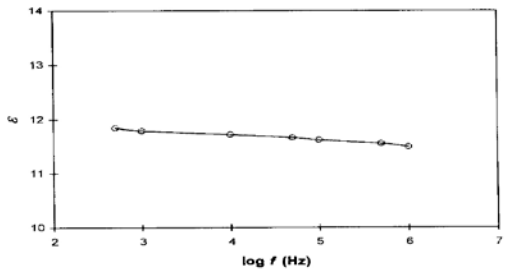


Fig.2 Variation of dielectric constant 'ε' with log frequency for pure GGG

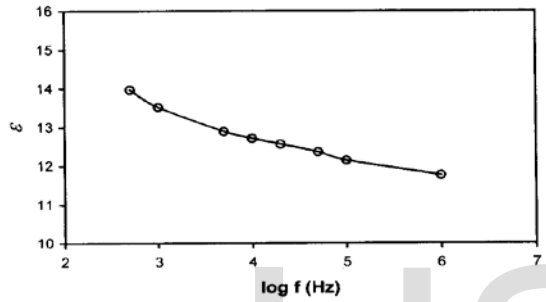


Fig.3 Variation of dielectric constant 'ε' with log frequency for GGG:Nd

The variation of dielectric loss (tanδ) with frequency for both samples is shown in figures 4 and 5 respectively. The variation in dielectric constant and dielectric loss is very small for pure GGG and are almost frequency independent, where as a slight increase in ε and tanδ are observed for Nd doped GGG with decreasing frequency.

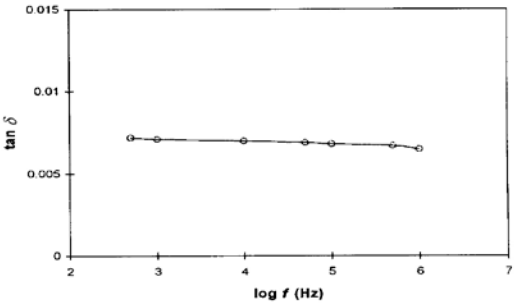


Fig.4 Variation of dielectric loss (tanδ) with log frequency for pure GGG

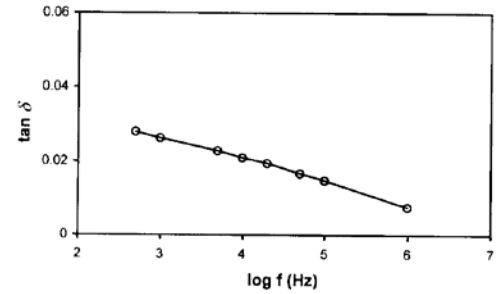


Fig.5 Variation of dielectric loss (tanδ) with log frequency for GGG:Nd

The room temperature dielectric constant and loss for both the samples at 1 MHz frequency are shown in table1.

TABLE 1

| Sample | Dielectric constant (ε) | Tanδ |
|---|---|---|
| GGG | 11.60 | $1.1 \times 10^{-3}$ |
| GGG:Nd | 11.76 | $2.3 \times 10^{-3}$ |

The variation of dielectric constant at different frequencies as a function of temperature is shown in figures 6 and 7 for both the crystals. A very little frequency dependent variation in ε is observed for pure GGG up to a temperature of 450$^0$C, which can be treated as almost constant with temperature and frequency. But above 450$^0$C temperature there is a considerable increase with temperature for 1 kHz and 10 kHz frequencies and a slow increase for higher frequencies. While for the Nd doped crystal there is a slow increase from room temperature to 400$^0$C, and is much rapid beyond this temperature, for all frequencies. Clear peaks in ε are observed at 500 Hz and 1 kHz for the doped crystal around 570$^0$C.

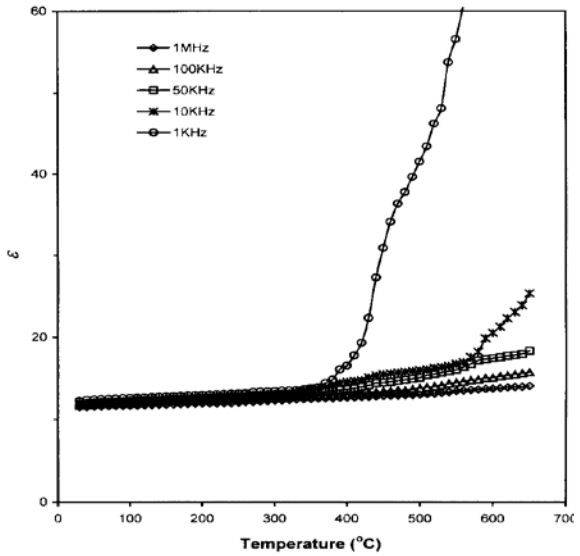


Fig.6 Variation of dielectric constant with temperature for pure GGG

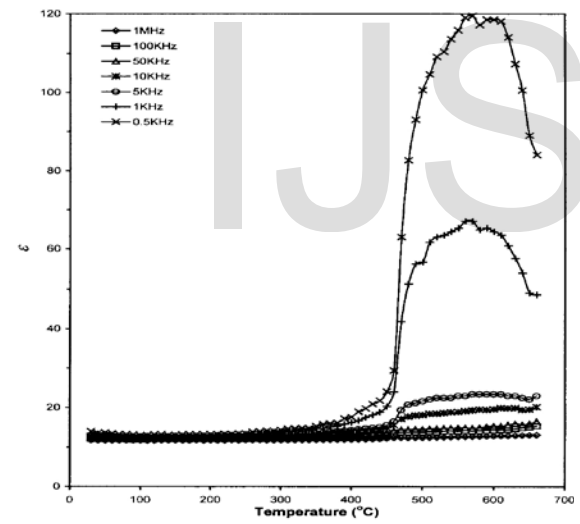


Fig.7 Variation of dielectric constant with temperature for GGG:Nd

The dielectric loss (tanδ) variation as a function of temperature for both the samples are shown in figures 8 and 9 respectively. For pure GGG crystal 'tanδ' is almost constant with temperature from room temperature to 450$^0$C and is found to be independent of frequency. Beyond 450$^0$C the loss increases with temperature, which is faster for low frequencies and slow for higher frequencies. For the Nd doped crystal dielectric loss is independent of frequency up to a temperature of 400$^0$C. Beyond this temperature it is frequency dependent and increases with increase of temperature. The increase in dielectric loss is low for higher frequency and a large increase is observed for all frequencies beyond 450$^0$C with a distinct peak around 620$^0$C. The changes observed in this region are similar to those observed in dielectric constant.

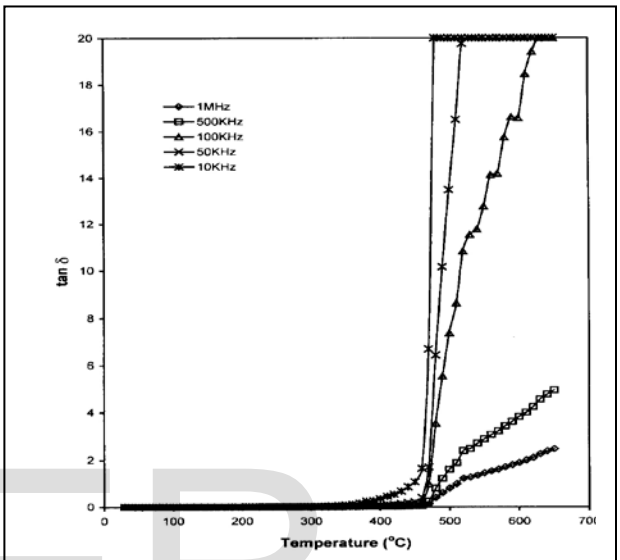


Fig.8 Variation of dielectric loss (Tanδ ) with temperature for GGG

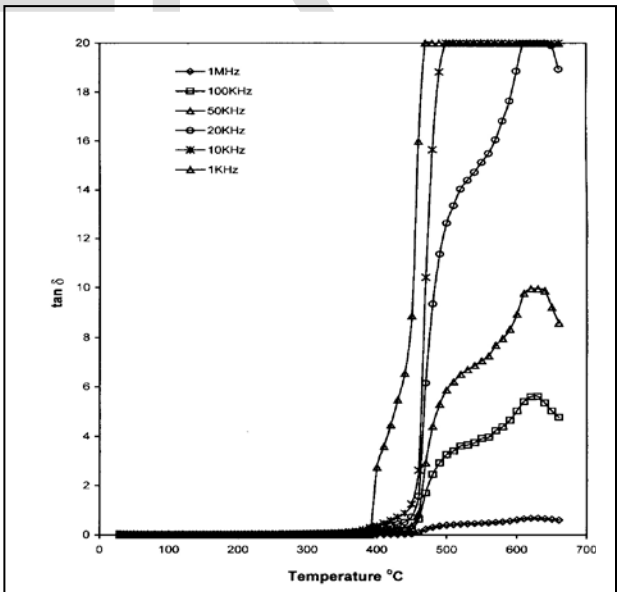


Fig.9 Variation of dielectric loss (Tanδ ) with temperature for GGG:Nd

The AC conductivity is calculated from the data on dielectric constant 'ε' and dielectric loss 'tanδ' using the Debye equation:

$$\sigma_{ac} = \varepsilon\varepsilon_0\omega \tan\delta$$

Where $\varepsilon_0$ is the permittivity of free space and ω is the angular frequency.

The variations of AC conductivity as a function of reciprocal temperature at various frequencies for the pure and Nd doped GGG samples are shown as Arrhenious plots in figures 10 and 11 respectively.

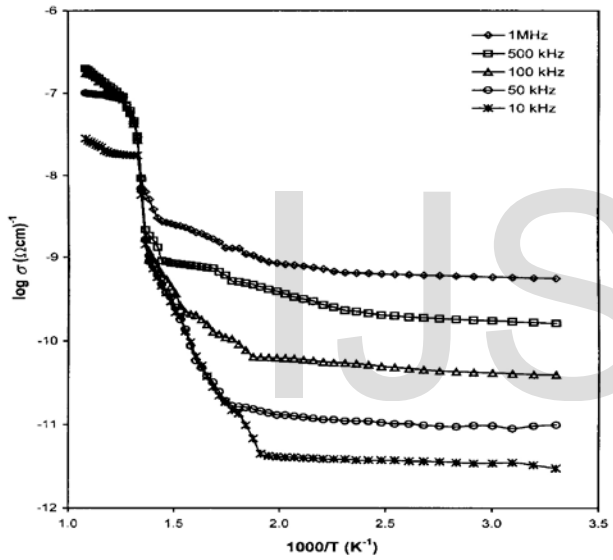


Fig.10 Variation of AC conductivity with reciprocal temperature for pure GGG

These plots show two regions. In the first region conductivity is almost constant up to about 400⁰C and a frequency dependent increase is observed at all temperatures. Beyond 400⁰C there is a steep and frequency independent increase in conductivity is observed.

## 4 DISCUSSION

The variation of dielectric constant and loss with frequency at room temperature shows the usual behaviour as observed for ionic crystals [2]. This is true for both the samples, pure and Nd doped GGG.

The variation with temperature shows distinct difference for the two samples, which is more pronounced for the variation of loss with temperature.

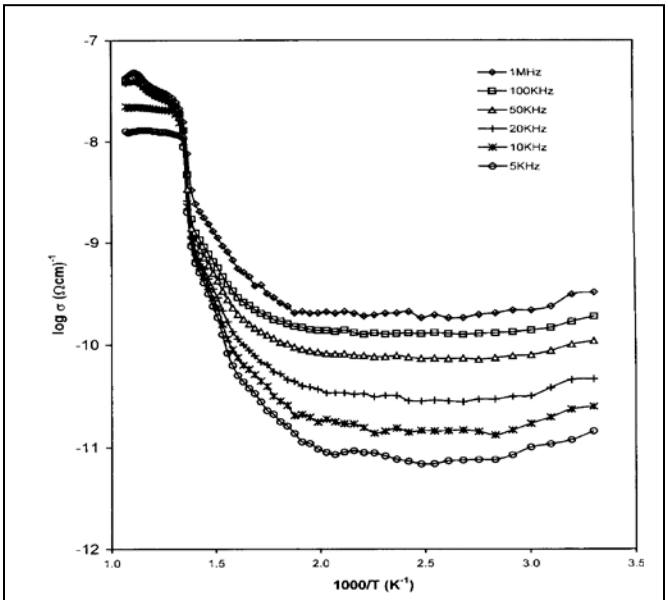


Fig.11 Variation of AC conductivity with reciprocal temperature for GGG:Nd

The changes at low frequencies is more prominent, while dielectric constant for the pure GGG sample shows a smooth variation up to a temperature of 450⁰C for all the frequencies a distinct sharp increase is observed for the Nd doped GGG sample. Again in the loss variation also while sharp increase is observed for the pure crystal, whereas for the Nd doped crystal the sharp increase is accompanied by a set of clear peaks around 620⁰C. Studies on luminescence by Andrichuk et. al. [3] have indicated that the intensity of luminescence emitted by GGG depends on the structural disorder. The fine structure absorption spectra (EXAFS) studied by Jun Dong and Kunquan Lu [4] has indicated the site exchange between Ga and Gd atoms in GGG and small displacements of atomic positions results in non cubic symmetry of garnets. The non-cubic symmetry is described by $R\bar{3}$. It is a small distortion from $Ia\bar{3}d$ space group. In the case of Nd doped Yttrium Aliminium Garnet ($Y_3Al_5O_{12}$:Nd) Bagdasarov et al. [5] have shown that the change in the structure is brought either by light pumping with UV radiation or by temperature. They have observed anomalies in dielectric constant (ε), loss (tanδ) and conductivity around 770K. These anomalies observed around 450⁰C and 630⁰C could be understood by comparing with the report on Nd doped Yttrium Aluminium Garnet [5]. For Nd:YAG the present authors also observed promising peaks in dielectric

constant and dielectric loss around $500^0$C for all frequencies [6]. They attribute this to a possible phase transition. The authors also observed a similar type of behaviour in the Nd and Cr co-doped GSGG [7]. In the present case also a similar type of changes are expected due to the presence of Nd in the c sub lattice.

Referring to the behaviour in the case of iron garnets, studied by the main author [8] the possible mechanism of conduction is by hopping of electrons. This hopping conduction is due to the exchange of electrons between divalent $Fe^{2+}$ and the trivalent $Fe^{3+}$ ions in YIG. But in the case of non-magnetic garnets such as GGG the exchange of electrons is not possible. Hence the possible mechanism is due to the motion of ions. The low value of conductivity at room temperature which of the order of $10^{-12}$ $\Omega^{-1}cm^{-1}$, also indicates the absence of electronic conduction. The possible charge carriers are the oxygen ions. The relaxation effects observed in tanδ could be explained as due to the dipole formed by the oxygen ion and $Nd^{3+}$ ion.

## 5 CONCLUSIONS

The dielectric behaviour of a pure and an Nd doped GGG single crystals are studied. Distinct peaks in ε and tanδ with temperature are observed for the Nd doped crystal which is due to a structural phase transition from $Ia\bar{3}d$ space group to $\bar{R}3$. The authors have observed that doping with Nd in a non-magnetic garnet brings a structural phase transition at a particular temperature which is characteristic of each garnet. The possible mechanism of conduction is by the motion of ions.

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