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# The Theoretical Investigation of the Impact of Substituting Bismuth Ion In Yttrium Iron Garnet (YIG) on The Faraday Rotation

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**Abstract:** By increasing information and communication technologies, the employment of linear polarized light during transmission through magnetic environments like Garnets has attracted attentions. In these environments we can significantly increase data capacitance and save speed as well as orbital switching speed through amplifying magneto optical interactions. According to experimental results, Yttrium Iron Garnet has higher magneto optical effects and replacing Bismuth partially by Yttrium ion, increases this effect greatly. Yet, there is no comprehensive theoretical analysis about this process. In this study, we try to present a logic reason for this process via the theoretical interpretation of light transmission in Yttrium-Iron Garnet and Bismuthic ones. The results of this study revealed that: a) the dominant magneto optical process in Yttrium-Iron Garnet in terms of crystal field transitions is paramagnetic transition and b) super-exchange field is increased in Bismuth Garnets due to the overlapping of Bismuth 6p orbital with Oxygen 2p orbital and Iron 3d orbital inhibiting the Zeeman splitting effect in stationary state. In addition, orbital overlapping increases spin -orbit coupling effect in excited state and causes energy levels to split. This kind of splitting is occurred due to diamagnetic charge transfer amplifying transferring as well as the Faraday Rotation Effect.

**Key words:** Magneto optical effect · Faraday rotation · Transmissions · $Y_3Fe_5O_{12}$ · $Y_{3-x}Bi_xFe_5O_{12}$

## INTRODUCTION

$RE_3Fe_5O_{12}$ is called Garnet due to the similarity of its crystal structure with that of a natural Garnet. It is written as follows considering its crystals sites [1]: $\{c^{3+}\}_3$ $[a^{3+}]_2$ $(d^{3+})_3O_{12}$.

In which a, d and c stand for cations which can occupy lattice positions with different oxygen coordination numbers as follows:

Dodecahedral with 8 oxygen around it at 24 positions, octahedral with 6 oxygen around it at 16 positions, d tetragonal with 4 oxygen around it at 24 positions and 96 positions for oxygen ions; totally form 160 ions within primitive cell. Similar to the primitive cell shown in Fig. 1, [a] octahedral and (d) tetragonal positions are occupied by iron or iron substitutions and {c} dodecahedral positions are occupied by rare earth cations or $La^{+3}$, $Y^{+3}$ and $Bi^{+3}$ ions [1].

Yttrium-Iron Garnet is one of the most important Garnets its specification is as follows composition $Y_3Fe_5O_{12}$, lattice constant: 12.376Å, volume per formula unit: 236.9Å$^3$, Curie temperature: $T_c$=559 K [2].

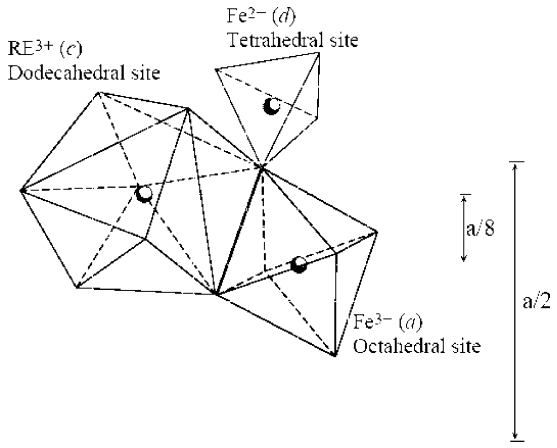


Fig. 1: The crystal structure of magnetic Garnets [1]

In Yttrium-Iron Garnet, 24 $Y^{+3}$ ions are distributed in c sites and 40 $Fe^{+3}$ ions are distributed in a and d sites. This is oxygen ion which makes these ions to interact with each other resulting in ferromagnetic state. The whole YIG is magnetized due to the resultant of two $Fe^{+3}$ sites which has been magnetized inverse directionally. In the absolute

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zero temperature, the share of each ion in magnetization process is $5\mu_b$ (Bohr magneton). Since within each formula two $Fe^{+3}$ ions, located on the (a) sites, are magnetized in the same direction and three $Fe^{+3}$ ions, located on the d sites, are magnetized inverse directionally, the resultant of each formula unit is 5 $\mu_b$. This substance is widely used in communication and light electronic industries due to its significant magneto optical properties as well as low losses and negligible absorption level in visible light and near to infra red ranges and it plays significant role in microwave filters, oscillators, modulators, light sensors and isolators [2]. In Garnet system, the most important magneto optical effect is the Faraday rotation i.e. the rotation of the polarization plane of linear polarized light during its transmission through a magnetic material in the presence of an external magnetic field. In anisotropic environments like Garnets, the Faraday rotation effect is generated due to the phase difference between the right hand rotating and the left hand rotating components of the linear polarized light. The origin of all this phenomena is optical transitions [3]. In addition to measuring possible transitions of this substance, Scott (Scott *et al.*) studied the transitions of Yttrium-Iron Garnet from the theoretical point of view [4]. Regarding the fact that iron has significant influence on the type of optical transitions in Yttrium-Iron Garnet, Borghi (Borghi *et al.*) analyzed the spectral properties of iron. According to the results of his analysis it was revealed that the type of optical transitions in this element is crystal field transition [5]. Moreover, in 1960 Buhrer declared that the Faraday rotation effect in Yttrium Iron Garnet can be increased by substituting Bismuth instates the some part of Bismuth [6]. In early 1980, it was reported that increasing Bismuth ions leads to a $\theta_f/x=2.06deg/\mu m$ (in which x is the number of Bismuth ions in a formula unit) linear increase in the Faraday rotation at the wavelength of 633 nm [3]. Since the ion radius of $Bi^{3+}$ which is equal to 1.123Å is higher than that of $Y^{+3}$, which is equal to 1.017Å, this increases the lattice constant from 12.37Å for YIG to 12.624Å for BIG. Scientific reports have approved a rise in the Curie temperature up to 34K due to the influence of Bismuth on super-exchange interactions. This confirms the increment of saturated magnetization in Bismuth Garnet [3]. According to Allen and Dionne (Allen & Dionne *et al.*) by replacing Bismuth the generated intense super-exchange field inhibits the Zeeman splitting in the stationary state and the general acceptable model consists only diamagnetic transitions [7]. According to Kahn's suggestion, in $Fe^{+3}$ ions the impermissible transitions of crystal fields may show paramagnetic magneto-optical effect while the transitions of permissible charge transfer may include diamagnetic effects [8]. Based on the mentioned issues, the object of this study is to theoretically study the impact of Bismuth ion on the increasing of the Faraday rotation effect in the various kinds of transitions in Bismuth Garnet compared with Yttrium-Iron Garnet (with no Bismuth).

**Description:** In classical approximation, energy varies continuously due to interactions between electrons and electric fields. But in the case that electrons, which have been constrained by core, have vibratory motion with the different values of $\omega$ due to magnetic forces, generated from external fields as well as spin-orbit coupling, the problem should be studied in the Quantum Mechanics environment [9]. In Garnets, magneto optic specification like the Faraday rotation lie inside a constant range of 1.5-5ev regarding refraction as well as light absorption indexes [10]. Therefore, these phenomena should be analyzed in terms of electron transition. The electron transition itself could be analyzed by studying the splitting generated due to crystal filed in metal cation levels located on octahedral or tetragonal positions or by the process of charge transfer from oxygen anion to $Fe^{+3}$ cation [9]. Considering the fact that within visible and infra red wavelengths, transitions are occurred due to vibration of electric dipoles, in anisotropic environments like Garnets magneto-optical properties are expressed by dielectric tensor. In dielectric tensor non diagonal components are based on electric dipoles and the oscillation intensity of these dipoles indicates the splitting range of energy levels ($\Delta$) which is the base of the Faraday rotation intensity [9]. Under this condition, there are two possible transitions in Garnets within infrared and visible wavelengths. By comparing the stationary state and the excited state as well as charge transferring quality the two mentioned transitions are divided into paramagnetic and diamagnetic classes. In the paramagnetic type, the Zeeman splitting is occurred due to applied field. It means that in these state ions get angular orbit momentum and after the exciting of energy levels they are degenerated [11] while in the diamagnetic class, the stationary state is singlet means that electron spins are degenerated and the super-exchange field removes this degeneration and in the excited state, levels are split due to spin-orbit coupling. In other words, ions have an orbital angular momentum in the excited state. These possible transitions can be expressed based on oscillation intensity as:

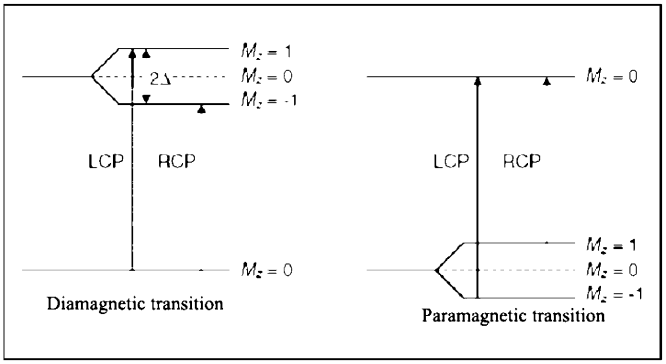


Fig. 2: Comparison of paramagnetic and diamagnetic transitions [12]

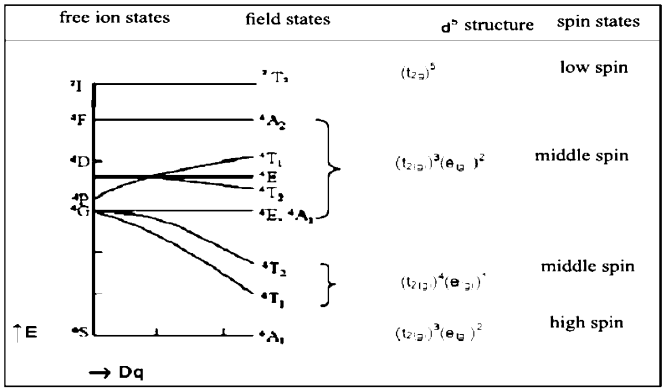


Fig. 3: Demonstration of the splitting of d5 orbital electron structure due to crystal field transition [13]

$$f = m\omega_o/h(<g|x|e>^2) \quad (1)$$

In which x is the operator of electric dipole which depends on spin-orbit coupling, <gl is the stationary state and le> is the excited state. In the paramagnetic transition, the right hand and the left hand rotating polarized lights have different oscillation intensity, f while in the diamagnetic transition, it is the same in both polarized elements. Fig. 2. shows a schematic view of the two transitions differences.

Paramagnetic transitions; comparing elliptical spectrum as well as the Faraday rotation with energy absorption band reveals that in $Fe^{+3}$ ion, the transitions of crystal fields are mostly paramagnetic transitions and in a regular magnetic system, field variations as well as spin-orbit coupling act strongly to generate a strong and effective field resulting in the forbidden paramagnetic transitions of electric dipoles, which depend on oscillating forces, as well as the right hand and the left hand rotating polarized light [3]. Thus, we should study the states of crystal filed in order to describe these transitions.

According to crystal field's electrostatic theory, the S spin of a free ion like $Fe^{+3}$ with the structure of $3d^5$ in a Ligand field can get three different states: full spin state (S=5/2,$^6$S) stationary state with sextet degeneration as well as middle spin exited state (S=3/2,$^4$G) with four degeneration and low spin excited state with double degeneration (S=1/2,$^2$I) [5]. Now, we study octahedral position in order to interpret crystal field's transitions based on the Ligand field of $Fe^{+3}$ ion. In octahedral position, the atomic orbitals of Fe ($3d^5$) are split into two orbitals groups: $t_{2g}$ and $e_g$ due to crystal field's transitions. The type of this splitting is (10Dq) considering crystal filed parameters. Regarding its energy level each orbital group is split again in the forms of high spin electrons (α spin) and low spin electrons) β spin). The positions through which the crystal fields can be transmitted have different electronic structure than $t_{2g}$ and $e_g$ orbitals.

Fig. 3. shows these possible transitions. According to this figure, the stationary state of $^6A_{1g}(^6S)$ with the structure of $(t^{\alpha}_{2g})^3(e^{\alpha}_{g})^2$ corresponds to Fe+3 full spin state and the first excited state $^4T_{1g}(^4G)$ with the structure of $(^{\alpha}t_{2g})^3(^{\beta}t_{2g})^1(^{\alpha}e_{1g})^1$ as well as $^4T_{2g}(^4G)$ with the structure of $(^{\alpha}t_{2g})^3(^{\beta}t_{2g})^1(^{\beta}e_{1g})^1$ corresponds to iron middle spin state all of them are split due to John Taylor effect.

Table 1: Paramagnetic optic transition parameters in YIG [14]

| Transition | (eV)hν | $\lambda_o$(nm) | $Fe^{3+}$ | Oscillation intensity |
|---|---|---|---|---|
| $^6A_{1g}(^6S)\rightarrow{}^4T_{1g}(^4G)$ | 1.38 | 900 | Octahedral | $2\times10^{-5}$ |
| $^6A_{1g}(^6S)\rightarrow{}^4T_{2g}(^4G)$ | 1.80 | 690 | Octahedral | $2\times10^{-5}$ |
| $^6A_{1g}(^6S)\rightarrow{}^4T_1\ (^4G)$ | 2.00 | 620 | Tetragonal | $8\times10^{-5}$ |
| $^6A_{1g}(^6S)\rightarrow{}^4T_2(^4G)$ | 2.44 | 510 | Tetragonal | $1.6\times10^{-4}$ |
| $^6A_{1g}(^6S)\rightarrow{}^4E,{}^4A_1\ (^4G)$ | 2.59 | 480 | Tetragonal | $3\times10^{-5}$ |
| $^6A_{1g}(^6S)\rightarrow{}^4E_g,{}^4A_{1g}\ (^4G)$ | 2.64 | 470 | Octahedral | $2\times10^{-5}$ |
| $^6A_{1g}(^6S)\rightarrow{}^4T_{2g}(^4D)$ | 2.82 | 440 | Octahedral | $1\times10^{-4}$ |
| $^6A_{1g}(^6S)\rightarrow{}^4T_2\ (^4D)$ | 3.03 | 410 | Tetragonal | $6\times10^{-5}$ |
| (CFT)diamagnetic transition | 2.89 | 430 | | $2\times10^{-3}$ |

Next excited states generating due to spin jump may convert to one of these structures: $(^{\alpha}t_{2g})^3(^{\beta}t_{2g})^1(^{\alpha}e_{1g})^2$ and $(^{\alpha}t_{2g})^3(^{\alpha}e_{1g})^1(^{\beta}e_{1g})^1$ and the double stationary state of $^2T_{2G}(^2I)$ corresponds to the stationary state with the low spin structure of $(t_{2g})^5$ [13].

Therefore, using critical factors like spin number, electron structure and energy variations, indicated in Fig. 3, it will be revealed that the minimum energy belongs to the full spin state with sextet degeneration and the first excited state generating due to crystal field splitting would be middle spin with higher energy and four degeneration and the final excited state would be low spin with the maximum energy and double degeneration. By this process we will obtain low spin state with higher energy through changing high spin state with lower energy. Thus, by applying an external field only the Zeeman splitting will be occurred which has the minimum level of energy. So, crystal field splitting matches with paramagnetic transition. According to Fig. 3 the minimum energy level, 0ev, belongs to $^6A_{1g}$ level and the transition from this level to other split levels, due to crystal field, is significantly important and the minimum transition in Garnet is in the energy level of hν=1.38 eV which corresponds to the wavelength of $\lambda_o$=900nm. It is related to $^6A_{1g}\rightarrow{}^4T_{1g}$ transition in octahedral position. Table 1 shows other transitions in YIG within each position in terms of wavelength, energy and oscillation intensity [14]. Therefore, according to the reported experimental and theoretical interpretations as well as Table 1 the most dominant property of magneto optic spectrum is the governing of paramagnetic transitions so that there is only one diamagnetic transition and the rest of them are paramagnetic. Thus, we can conclude that the dominant transition in Yttrium-Iron Garnet is paramagnetic transition [15].

**Diamagnetic Transitions:** Regarding rotation interpretation and the elliptical spectrum of Yttrium-iron Garnet and according to the diamagnetic functions of the non diagonal components of electric susceptibility tensor i.e. $\epsilon_1=\epsilon'_1+i\epsilon''1$ the equation of this type of transition is written as follows [9]:

$$\epsilon_1=(\omega^2{}_p\Sigma^{-}{}_{+}\pm(f_{\pm}/2\omega_{o\pm}))\times(\omega(\omega^2{}_{o\pm}-\omega^2-\Gamma^2)+i\Gamma(\omega^2{}_{o\pm}+\omega^2+\Gamma^2))/((\omega^2{}_{o\pm}-\omega^2+\Gamma^2)^2+4\omega^2\Gamma^2 \quad (2)$$

In which $\omega^2{}_p=4\pi Ne^2/m$ is the square of plasma frequency, e and m are Electron charge and electron mass respectively and n is the number of $Fe^{3+}$ ions within each position in cm3. Also,we have: ωo±=ωo±Δ in which ω0 is transition frequency and Δ is the splitting occuring in diamagnetic exciting state. f± = (f/2) (1±Δ/ω0) is the intensity of positive and negative mode's oscillation and Γ is the specification of half width of transition line. Also, we can write the Faraday rotation angle in terms of dielectric tensor's non diagonal components as follows:

$$\theta F\tilde{}\ (\omega/2nc)\epsilon'1 \quad (3)$$

Regarding diamagnetic functions properties i.e. the dependency of electrical susceptibility as well as oscillation intensity on the splitting level of Δ we can conclude that in diamagnetic transition the most important parameter which helps us to obtain significant Faraday rotation is the splitting level of Δ. This dependency is completely appropriate for a transition in $\omega\approx\omega_0$ condition and regarding diamagnetic function, $\varepsilon_1$ is completely proportional with Δ. Therefore, the elements of $\varepsilon_1$ is linear by Δ in the first order. Thus, increasing Δ increases the Faraday rotation tangibly. In diamagnetic transitions inside Garnets, the splitting level of Δ is determined through the value of spin-orbit coupling occurring due to $Fe^{+3}$ electron valences. Regarding the regularity of electron's spins this coupling is occurred when an applied filed could overcome the variations of the internal field of substances [16].

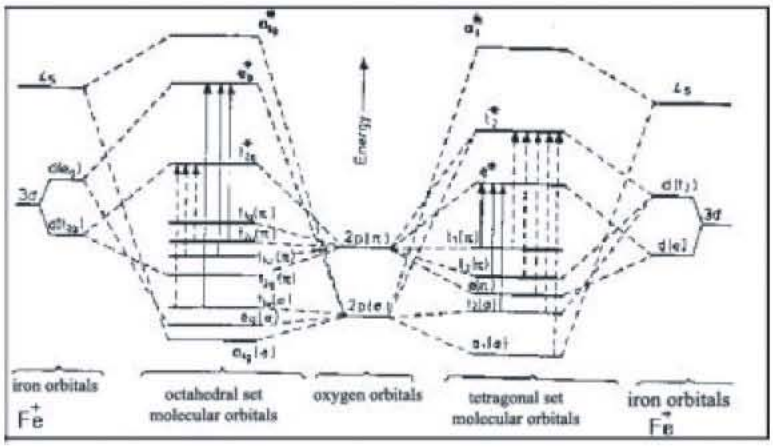


Fig. 4: Molecular orbitals of $FeO_6$ octahedral positions and $FeO_4$ tetragonal positions [10]

In order to make relation between orbital splitting and spin-orbital coupling in Garnets, we can determine the permissible transitions of electric dipoles based on Molecular Orbital Theory. Considering Garnet's molecular structure which includes octahedral and tetragonal positions each of them has $O^{2-}$ ion and $Fe^{3+}$ cation. Also, based on the molecular orbital theory we can expect the optical activity of YIG, relating to the oxygen- ions of octahedral and tetragonal positions, due to overlapping of oxygen 2p orbital and iron ion 3d and 4s orbitals.

Moreover, in these positions dipoles transitions are charge transfer type in which one electron from the p orbital of oxygen is transferred to the main anti bond orbital of metal ion. Therefore, we study only transitions to the lowest anti bond orbital which has $e^*_g$ or $t^*_{2g}$ symmetry in octahedral positions and $e^*$ or $t^*_2$ symmetry in tetragonal positions (they are highlighted by star).

Permissible charge transfers with high magneto optic influence on this composition include: $t^n_{2u}(\pi)$, $t_{1u}(\pi)$, $t_{1u}(\sigma)\rightarrow t^*_{2g}$ and $t^n_{2u}(\pi)$, $t_{1u}(\pi)$, $t_{1u}(\sigma)\rightarrow e^*_g$ for octahedral positions as well as ($t^n_{2u}(\pi)$, $t_{1u}(\pi)$, $t_{1u}(\sigma)\rightarrow t^*_2$ and ($t^n_{2u}(\pi)$, $t_{1u}(\pi)$, $t_{1u}(\sigma)\rightarrow e^*$ for tetragonal positions. The sum of these transitions is 12 which have been shown by staff lines in Fig. 4. [8].

$a^*_{1g}$, $a^*_1$, $e^*$ and $e^*_g$ orbitals have no orbital momentum due to their symmetry, therefore when we investigate optical transitions in YIG, we should pay attention to the transitions to $t^*_{2g}$ and $t^*_2$ shown in figure by dashed lines [10]. When we introduce Bismuth to garnet lattice, we should expect two possible orbitals bonds: the bond of Bismuth ion 6S and 6P orbitals with adjacent oxygen 2p valence orbital; and the bond of Bismuth ion 6S and 6P orbitals with iron 3d orbital via intermediate oxygen orbitals. Based on theoretical studies as well as experimental reports, interactions between Bismuth orbital and oxygen and iron orbitals would give two important results: a) the oscillation intensity of transitions has been increased in the new composition compared with YIG. It means that those transitions which showed no Faraday rotation in YIG, show it in Bi:YIG due to amplification. Similar to the transitions to the end states of $e^*$and $e^*_g$ in YIG in which spin-orbital coupling coefficient, $\xi$, was zero due to symmetry in Bismuth Garnet, the symmetry are eliminated and the value of this coefficient would be non zero. Moreover, these transitions are done in BIG at low levels of energy (2.6eV) compared with transition energy in YIG (4eV). Transitions like $t_{1u}(\pi)$, $t_{2u}(\pi)\rightarrow e^*$ in tetragonal positions which are done due to orbital interactions between oxygen 2p orbital and Bismuth 6p orbital at low levels of energy is an evidence. But in orbitals like $a_1(\sigma)$ even during interaction between them and Bismuth ions the value of spin-orbital coupling remains zero since the symmetry is not disturbed [17]. b) the higher coefficient of spin-orbital coupling in Bismuth 6P electrons compared with free ion, is a cause which increases spin-orbital coupling in interactions between oxygen and iron. This cause the Faraday rotation to increase in Bi:YIG by 100 times compared with YIG at the wavelength of 633nm. The origin of these increases is the hybridizing of Bismuth orbital with oxygen 2P orbital as well as iron 3d orbital [17]. It should be mentioned that the substitution of Bismuth is not considered as a direct cause for magneto optic transitions as well as significant increase of the Faraday rotation. These transitions are not occurred within Bismuth ions and the ions only amplify small transitions in Garnets with no Bismuth and cause them to appear [6]. Considering the fact that increasing Bismuth ion in ferromagnetic Garnets increases magneto

optic properties due to amplifying some transitions, we can argue that besides the interactions of $O^{2-}$ and $Fe^{3+}$, the rare soil ions located on dodecahedral positions can increase the Faraday rotation. Also, this increase may occur due to the influence of Bismuth on iron's sub lattices which increases transitions through strong changes interactions between $Fe^{3+}_{tet}$ and $Fe^{3+}_{oct}$ or due to internal charge transfer transitions increase [12].

According to the molecular orbital theory we expected 14 possible charge transfer transitions at each sub lattices of YIG but practically there were appeared no magneto optic effect while in Bismuth Garnet these transitions showed high magneto optic effects at visible and infra red wavelengths due to orbital spin interactions between Bismuth 6P orbital and oxygen 2P orbital as well as iron 3d orbital at low levels of energy [18]. Based on theoretical computations these spectral variations are generated due to four new type $\pi$ charge transfer transition (diamagnetic transitions) from $2P(O^{2-})$ to $3d(Fe^{3+})$ including $t_{1u}(\pi)$, $t_{2u}(\pi) \rightarrow t^*2g$ transitions in octahedral positions and $t_{1u}(\pi)$, $t_{2u}(\pi) \rightarrow e^*$ transitions in tetragonal positions [18]. Thus, replacing bismuth ion by Yttrium ions increases exchange interactions between iron positions and increases charge transfer from oxygen anion to iron cation due to the overlapping of Bismuth orbital with oxygen and iron orbitals. This in turn will lead to the increasing of super exchange field in the stationary state and inhibits the Zeeman splitting effect in this state. The charge transfer from the stationary state to the excited state increases spin-orbital coupling in the excited state and causes the orbitals of energy levels to split. Therefore, we can argue that in Bismuth Garnet the charge transfer transitions are diamagnetic.

## CONCLUSION

- Considering electron exchanges between $O^{2-}$ and $Fe^{+3}$ orbital in YIG, increasing Bismuth increases charge transfer between $O^{2-}$ and $Fe^{+3}$ orbital. This in turn increases the intensity of transitions in tetrahedral and octahedral positions compared with previous state. The transitions of octahedral positions have the main share in magneto optic effects.
- Since in Bismuth Garnet, transitions as well as spin-orbital coupling are increased, we can argue that some transitions which showed no magneto optical effects in Garnet with no Bismuth, due to negligible spin-orbital interactions, this time showed the effect due to the increase of spin-orbit interaction at lower energy levels.
- According to theoretical studies as well as reported experimental results, in YIG the dominant transitions are paramagnetic crystal field transitions while form practical and theoretical points of view the dominant transitions in Bi-YIG are diamagnetic charge transfer transitions. So, in Bismuth Garnet the optical processes are diamagnetic processes.
- In paramagnetic transitions the oscillation intensity of electric dipoles is not the same in the right hand rotating and the left hand rotating polarized light while in the diamagnetic transitions the intensity parameter is the same in the both polarized lights because the amount of absorption is neglegible. For this, the Faraday rotation speed increases in diamagnetic transition compared with paramagnetic one so that the type of transition in Bismuth Garnet is diamagnetic.
- As a hypothesis it could be said that magnetic environments with diamagnetic transitions as their dominant transitions have higher magneto optical effects compared with the environments with paramagnetic transitions as their dominant transition. Therefore, the Faraday rotation would be higher in the former than the latter.

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