A First Principle Study on Magneto-Optical Effects and Magnetism in Ferromagnetic Semiconductors $Y_3Fe_5O_{12}$ and $Bi_3Fe_5O_{12}$

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The magneto-optical (MO) effects not only are a powerful probe of magnetism and electronic structure of magnetic solids but also have valuable applications in high-density data-storage technology. Yttrium iron garnet ($Y_3Fe_5O_{12}$) (YIG) and bismuth iron garnet ($Bi_3Fe_5O_{12}$) (BIG) are two widely used magnetic semiconductors with strong magneto-optical effects and have also attracted the attention for fundamental physics studies. In particular, YIG has been routinely used as a spin current injector. In this paper, we present a thorough theoretical investigation on magnetism, electronic, optical and MO properties of YIG and BIG, based on the density functional theory with the generalized gradient approximation plus onsite Coulomb repulsion. We find that both semiconductors exhibit large MO effects with their Kerr and Faraday rotation angles being comparable to that of best-known MO materials such as MnBi. Especially, the MO Kerr rotation angle for bulk BIG reaches -1.2° at photon energy $\sim 2.4$ eV, and the MO Faraday rotation angle for BIG film reaches -74.6 °/µm at photon energy $\sim 2.7$ eV. Furthermore, we also find that both valence and conduction bands across the MO band gap in BIG are purely spin-down states, i.e., BIG is a single spin semiconductor. These interesting findings suggest that the iron garnets will find valuable applications in semiconductor MO and spintronic nanodevices. The calculated optical conductivity spectra, MO Kerr and Faraday rotation angles agree well with the available experimental data. The main features in the optical and MO spectra of both systems are analyzed in terms of the calculated band structures especially by determining the band state symmetries and the main optical transitions at the $\Gamma$ point in the Brillouin zone.

I. INTRODUCTION

Yttrium iron garnet ($Y_3Fe_5O_{12}$, YIG) is a ferrimagnetic semiconductor with excellent magnetic properties such as high curie temperature $T_c$ [1], low Gilbert damping $\alpha \sim 6.7 \times 10^{-5}$ [2-4] and long spin wave propagating length [5]. Various applications such as spin pumping require a non-metallic magnetic material. YIG is thus routinely used for spin pumping purposes [4]. It is also widely used as a magnetic insulating substrate for purposes such as introducing magnetic proximity effect while avoiding electrical short-cut. YIG has high Curie temperature, which is good for applications across a wide temperature range. The low Gilbert damping of YIG also makes it a good microwave material. YIG thus becomes a famous material in the field of spintronics, where coupling between magnetism, microwave and spin current becomes possible.

Magneto-optical (MO) effects are important examples of light-matter interactions in magnetic phases. [6, 8] When a linear polarized light beam is shined from a dielectric to a magnetic material, the reflected and transmitted light becomes elliptically polarized. The principle axis is rotated with respect to the polarization direction of incident light beam. The former and latter effects are termed MO Kerr (MOKE) and MO Faraday (MOFE) effects, respectively. MOKE allows us to detect the magnetization locally with a high spatial and temporal resolution in a non-invasive fashion. Furthermore, magnetic materials with large MOKE would find valuable MO storage and sensor applications [6, 10]. Thus it has been widely used to probe the electronic and magnetic properties of solids, surface, thin films and 2D magnets [8]. On the other hand, MOFE can be used as a time-reversal symmetry-breaking element in optics [11], and its applications such as optical isolators are consequences of time-reversal symmetry-break [12]. Magnetic materials with large MOKE or MOFE rotation angles have technological applications.

YIG is also known to be MO active [13]. Various experiments have been carried out to study the MOKE and MOFE of iron garnets in the visible and near-UV regime [14, 15]. Substituting yttrium with bismuth result in bismuth iron garnet ($Bi_3Fe_5O_{12}$) (BIG). BIG has approximately 7 times larger Faraday rotation angles than that of YIG. The effect of doping bismuth into YIG on the MOFE of iron garnets in the visible and near-UV regime [14, 15]. Using first principle calculation on the MOKE or MOFE spectra of...
YIG and BIG has been reported. Therefore, here we carry out a systematic first-principle density functional study on the optical and MO properties of YIG and BIG. The rest of this paper is organized as follows. A brief description of the crystal structures of YIG and BIG as well as theoretical methods is given in Sec. II. In Sec. III, the calculated magnetic moments, electronic structure, optical conductivities, MO Kerr and Faraday effects are presented. Finally, the conclusions drawn from this work are given in section IV.

II. CRYSTAL STRUCTURE AND COMPUTATIONAL METHODS

Bulk YIG and BIG crystalize in the cubic structure with space group Ia3d [21, 22], as illustrated in Fig. 1(a). In each unit cell, there are 48 oxygen atoms at the Wyckoff 96h positions, 8 octahedrally coordinated iron atoms (Fe\(^{O}\)) at the 16a positions, and 12 tetrahedrally coordinated iron atoms (Fe\(^{T}\)) at the 24d positions in the primitive cell. In other words, there are two Fe\(^{O}\) ions and three Fe\(^{T}\) ions per formula unit (f.u.). The experimental lattice constant \(a = 12.376\) Å, and the experimental Wyckoff parameters for oxygen atoms are \((x, y, z) = (0.9726, 0.0572, 0.1492)\). [21] Substituting yttrium with bismuth in YIG results in BIG. The experimental lattice constant for BIG \(a = 12.6469\) Å. [22] Accurate oxygen position measurement for BIG is still on demand and under debate [18]. Therefore we use the experimental parameters for all YIG calculations, while the experimental lattice constant is used for BIG with the atomic positions determined theoretically (see Table I).

Our first principle calculations are based on the density functional theory with the generalized gradient approximation (GGA) of the Perdew-Burke-Ernzerhof formula [22] to the electron exchange-correlation potential. Furthermore, we use the GGA + \(U\) method to have a better description for on-site interactions for Fe \(d\) electrons. [24] Here we set \(U = 4.0\) eV, which was found to

TABLE I. The structural parameters of Y\(_3\)Fe\(_5\)O\(_{12}\) and Bi\(_3\)Fe\(_5\)O\(_{12}\) used in this work. For YIG, experimental lattice constant \(a = 12.376\) Å and oxygen positions [21] are used. For BIG, experimental lattice constant \(a = 12.6469\) Å [22] is used while the oxygen positions are determined theoretically.

| Y\(_3\)Fe\(_5\)O\(_{12}\) Wyckoff position | \(x\) | \(y\) | \(z\) |
|--------------------------|------|------|------|
| Fe\(^{O}\) 16a           | 0.0000 | 0.0000 | 0.0000 |
| Fe\(^{T}\) 24d           | 0.3750 | 0.0000 | 0.2500 |
| Y 24c                  | 0.1250 | 0.0000 | 0.2500 |
| O 96h                  | 0.9726 | 0.0572 | 0.1492 |

| Bi\(_3\)Fe\(_5\)O\(_{12}\) Wyckoff position | \(x\) | \(y\) | \(z\) |
|--------------------------|------|------|------|
| Fe\(^{O}\) 16a           | 0.0000 | 0.0000 | 0.0000 |
| Fe\(^{T}\) 24d           | 0.3750 | 0.0000 | 0.2500 |
| Bi 24c                  | 0.1250 | 0.0000 | 0.2500 |
| O 96h                  | 0.0540 | 0.0300 | 0.1485 |

We first calculate the optical conductivity tensor which can be written in the following form [29]:

\[
\sigma = \begin{pmatrix}
\sigma_{xx} & \sigma_{xy} & 0 \\
-\sigma_{xy} & \sigma_{xx} & 0 \\
0 & 0 & \sigma_{zz}
\end{pmatrix}.
\] (1)

The optical conductivity tensor can be formulated within...
the linear response theory. Here the real part of the diagonal elements and imaginary part of the off-diagonal elements are given by \[29 31.\]

\[
\sigma_{aa}^1(\omega) = \frac{\pi e^2}{\hbar \omega_0^2} \sum_{i,j} \int_{BZ} \frac{dk}{(2\pi)^3} |p_{ij}^n|^2 \delta(\epsilon_{kj} - \epsilon_{ki} - \hbar \omega),
\]

(2)

\[
\sigma_{xy}^2(\omega) = \frac{\pi e^2}{\hbar \omega_0^2} \sum_{i,j} \int_{BZ} \frac{dk}{(2\pi)^3} \text{Im}[p_{ij}^x p_{ij}^y] \delta(\epsilon_{kj} - \epsilon_{ki} - \hbar \omega),
\]

(3)

where \( \hbar \omega \) is the photon energy, and \( \epsilon_{ki(j)} \) are the energy eigenvalues of occupied (unoccupied) states. The transition matrix elements \( p_{ij}^n = \langle kj | \hat{p}_a | ki \rangle \) where \( | ki(j) \rangle \) are the \( i(j) \)th occupied (unoccupied) states at \( k \)-point \( k \), and \( \hat{p}_a \) is the Cartesian component \( a \) of the momentum operator. The imaginary part of the diagonal elements and the real part of the off-diagonal elements are then obtained from \( \sigma_{aa}^1(\omega) \) and \( \sigma_{xy}^2(\omega) \), respectively, via the Kramers-Kroons transformations as follows:

\[
\sigma_{aa}^1(\omega) = -\frac{2\omega}{\pi} P \int_0^\infty \frac{\sigma_{aa}^1(\omega')}{\omega' - \omega^2} d\omega',
\]

(4)

\[
\sigma_{xy}^2(\omega) = \frac{2\omega}{\pi} P \int_0^\infty \frac{\sigma_{xy}^2(\omega')}{\omega' - \omega^2} d\omega',
\]

(5)

where \( P \) denotes the principle value of the integration. We can see that Eq. (2) and Eq. (3) neglect transitions across different \( k \)-points since the momentum of the optical photon is negligibly small compared with the electron crystal momentum and thus only the direct interband transitions need to be considered. In our calculations \( p_{ij}^n \) are obtained in the PAW formalism \[32.\] We use a \( 10 \times 10 \times 10 \) \( k \)-point mesh and the Brillouin zone integration is carried out with the linear tetrahedron method (see \[33.\] and references therein), which leads to well converged results. To ensure that the \( \sigma_{aa}^2(\omega) \) and \( \sigma_{xy}^1(\omega) \) in the optical frequency range (e.g., \( \hbar \omega < 8 \text{ eV} \)) obtained via Eqs. (4) and (5) are converged, we include the unoccupied states at least 21 eV above the Fermi energy, i.e., a total of 1200 (1300) bands are used in the YIG (BIG) calculations.

For a bulk magnetic material, the complex polar Kerr rotation angle is given by \[34 35.\]

\[
\theta_K + i \epsilon_K = \frac{-\sigma_{xy}}{\sigma_{xx} \sqrt{1 + i(4\pi/\omega) \sigma_{xx}}},
\]

(6)

Similarly, the complex Faraday rotation angle for a thin film can be written as \[36.\]

\[
\theta_F + i \epsilon_F = \frac{\omega d}{2c} (n_+ - n_-),
\]

(7)

where \( n_+ \) and \( n_- \) represent the refractive indices for left- and right-handed polarized lights, respectively, and are related to the corresponding dielectric function (or optical conductivity via expressions \( n_\pm^2 = \varepsilon_\pm = 1 + \frac{4\pi}{\omega} \sigma_\pm = 1 + \frac{4\pi}{\omega} (\sigma_{xx} \pm i \sigma_{xy}) \). Here the real parts of the optical conductivity \( \sigma_\pm \) can be written as

\[
\sigma_\pm(\omega) = \frac{\pi e^2}{\hbar \omega_0^2} \sum_{i,j} \int_{BZ} \frac{dk}{(2\pi)^3} \Pi_{ij}^\pm |^2 \delta(\epsilon_{kj} - \epsilon_{ki} - \hbar \omega),
\]

where \( \Pi_{ij}^\pm = \langle kj | \hat{p}_a \pm i \hat{p}_b | ki \rangle \). Clearly, \( \sigma_{xy} = \frac{1}{\sqrt{2}} (\sigma_+ - \sigma_-) \), and this shows that \( \sigma_{xy} \) would be nonzero only if \( \sigma_+ \) and \( \sigma_- \) are different. In other words, magnetic circular dichroism is the fundamental cause of the nonzero \( \sigma_{xy} \) and hence the MO effects.

### III. RESULTS AND DISCUSSION

#### A. Magnetic moments

Here we first present calculated total and atom-decomposed magnetic moments in Table I. As expected, \( Y_3\text{Fe}_5\text{O}_{12} \) is a ferrimagnet in which Fe ions of the same type couple ferromagnetically while Fe ions of different types couple antiferromagnetically, as illustrated in Fig. 1(b). Since there are two \( \text{Fe}^5 \) ions and three \( \text{Fe}^7 \) ions in a unit cell, \( Y_3\text{Fe}_5\text{O}_{12} \) is ferrimagnetic with a total magnetic moment per f.u. being \( \sim 5.0 \mu_B \) (see Table I). The calculated spin magnetic moments of Fe ions of both types are \( \sim 4.0 \mu_B \), being consistent with the high spin state of Fe ions. For \( Y_3\text{Fe}_5\text{O}_{12} \), the orbital magnetic moments of Fe are parallel to their spin magnetic moments. Nonetheless, the calculated orbital magnetic moments of Fe are small, because of strong crystal field quenching. Interestingly, there is a significant spin magnetic moment on each O ion, and this together with the spin magnetic moment of one net Fe ion per unit cell leads to the total spin magnetic moment per f.u. of \( \sim 5.0 \mu_B \). The calculated Fe magnetic moments for both symmetry sites agree rather well with the measured ones of \( \sim 4.0 \mu_B \). \[37.\] The calculated total magnetization of \( \sim 5.0 \mu_B/\text{f.u.} \) is also in excellent agreement with the experiment. \[37.\]

\( \text{Bi}_3\text{Fe}_5\text{O}_{12} \) is also predicted to be ferrimagnetic, although the calculated magnetic moments of both \( \text{Fe}^5 \) and \( \text{Fe}^7 \) ions are slightly smaller than the corresponding ones in \( Y_3\text{Fe}_5\text{O}_{12} \) (see Table I). The total magnetization and local magnetic moments of other ions in BIG are almost identical to that in YIG. However, the experimental \( m_{\text{tot}} \) for BIG is only \( 4.4 \mu_B \), \[38.\] being significantly smaller than the calculated value. As mentioned before, stable high quality BIG crystals are hard to grow. Consequently, this notable discrepancy in total magnetization between the calculation and the previous experiment \[38.\] could be due to the poor quality of the samples used in the experiment.
TABLE II. Total spin magnetic moment ($m^t$), atomic (averaged) spin magnetic moments ($m^F_e$, $m^O_s$, $m^Y_{(Bi)}$), atomic (averaged) Fe orbital magnetic moments ($m^{Fe}_{o}$) and band gap ($E_g$) of ferrimagnetic Y$_3$Fe$_5$O$_{12}$ and Bi$_3$Fe$_5$O$_{12}$ from the full-relativistic electronic structure calculations. For comparison, the available measured optical $E_g$ and total magnetization $m^t_{exp}$ are also listed.

| Structure   | $m^t$ ($m^t_{exp}$) ($\mu_B$/f.u.) | $m^F_e$($16a$) ($m^F_e$($16a$)) ($\mu_B$/atom) | $m^F_e$($24d$) ($m^F_e$($24d$)) ($\mu_B$/atom) | $m^O_s$ ($\mu_B$/atom) | $m^Y_{(Bi)}$ ($\mu_B$/atom) | $E_g$ ($E_g^{exp}$) (eV) |
|-------------|----------------------------------|---------------------------------------------|---------------------------------------------|----------------|-----------------|----------------|
| Y$_3$Fe$_5$O$_{12}$ | 4.999 (5.0$^a$) | -4.177 (-0.016) | 4.075 (0.018) | 0.067 | 0.005 | 1.81 (2.4$^b$) |
| Bi$_3$Fe$_5$O$_{12}$ | 4.996 (4.4$^c$) | -4.161 (-0.018) | 4.068 (0.019) | 0.066 | 0.005 | 1.82 (2.1$^d$) |

$^a$Ref. 37. $^b$Ref. 14. $^c$Ref. 38. $^d$Ref. 17.

FIG. 2. Spin-polarized density of states of Y$_3$Fe$_5$O$_{12}$ from the scalar-relativistic calculation.

FIG. 3. Spin-polarized density of states of Bi$_3$Fe$_5$O$_{12}$ from the scalar-relativistic calculation.

**B. Electronic structure**

Here we present the calculated scalar-relativistic band diagrams of YIG and BIG. A suitable value of $U = 4$ eV is used since better agreements with experimental measurements are obtained in magnetism, optical, and MO spectrums. Our band diagram calculations show that YIG and BIG are both direct band-gap semiconductors, where the conduction band minimum (CBM) and valence band maximum (VBM) are both located at the Γ point. For BIG, CBM and VBM are both pure spin-up bands. This means that BIG is a single-spin semiconductor, which may find applications for efficient spintronic and spin photovoltaic devices. The calculated band gaps are shown in Table 1. Note that for YIG, we still underestimate band gap even when GGA+U method is used. The reason is due to that the lowest conduction bands at $E = 1.8 \sim 2.5$ eV above VBM are not accessible with optical techniques since very few states are in this energy range and cannot contribute much to optical transitions. The first main absorption edge for YIG is $\sim 2.5$ eV above VBM in our calculation, corresponding to first main absorption edge $\sim 2.4$ eV in experimental optical spectrums [14]. Since the origin of MO effects in solid is
FIG. 4. (a) Scalar-relativistic spin-polarized band structure and (b) fully relativistic band structure of $Y_3Fe_5O_{12}$.

due to magnetic dichorism (Eq. (8)), which can not happen without the presence of spin-orbit coupling. It will be interesting to examine how does spin-orbit coupling influence the band diagram. The full relativistic band diagrams for YIG and BIG are also presented in Fig. 4(b) and Fig. 5(b). As one can see, the YIG band diagram does not change significantly after considering spin-orbit coupling, while the BIG band diagram is drastically different. The gap at $\sim 3.9$ to $\sim 4.5$ eV above Fermi energy was at $\sim 3.4$ to $\sim 3.7$ eV above Fermi energy without considering spin-orbit coupling. This suggest that the effect of spin-orbit coupling on band diagram for BIG cannot be approximated as a perturbation, while that for YIG can. The change for substituting bismuth with yttrium not only enhanced the spin-orbit coupling but also changed the electronic structure drastically. We also calculated total as well as site-, orbital-, and spin-projected densities of states (DOS) for YIG and BIG. One can see partial DOS for Y dominates at $E > 4$ eV. Previous first principle works suggest the strong MO effects in BIG are due to the strong spin-orbit coupling (SOC) introduced by Bi atoms in BIG, and that Bi orbitals mainly
mix with conduction states since they energetically overlap with Bi orbitals. Here we reconfirm their result in the case of YIG(BIG) for yttrium(bismuth) orbitals mainly mix with the conduction band. The bismuth s orbitals are in the energy range of valence band, however due to its closed-shell nature we do not expect bismuth s orbitals to mix with iron or oxygen orbitals in the valence band. As the valence band energy dispersion looks alike with or without SOC in energy range $-2 \sim 0$ eV, this also suggest that bismuth s orbitals does not introduce SOC to valence band through mixing. From the partial DOS, we can see that for the BIG DOS spectrum in the scalar-relativistic approximation, there are bismuth orbitals near the gap at $\sim 3.4$ to $\sim 3.7$ eV. One can consider the effect of spin-orbit coupling as a gradually turned on interaction, the gap thus opens during the process due to the presence of bismuth orbital(FIG. 5.). This is the reason why the BIG band diagram changed drastically with the presence of spin-orbit coupling, as bismuth orbital is mixed with the conduction bands from $\sim 2$ eV to $\sim 6$ eV. On the other hand, the yttrium partial DOS mostly lie above 4.2 eV in the YIG DOS spectrum. The yttrium partial DOS looks more localized in energy domain compare to that of bismuth partial DOS. And the yttrium orbital does not contribute much to the partial DOS below the gap at $\sim 4$ eV to $\sim 4.2$ eV. This suggest that the yttrium orbital does not mix with the Fe and O in the conduction band of YIG as much of bismuth orbitals do in the case of BIG. For the partial DOS spectrums for Fe$^T$ and Fe$^O$ atoms, both takes similar features in the case of YIG and BIG. The Fe$^T$ partial DOS in the conduction band is fully spin down, while the Fe$^O$ partial DOS in the conduction band is fully spin up. The DOS peak a is mostly consists of t$_{2g}$ orbital while peak b is mostly consists of e$_g$ orbital, therefore the formulation of the gap for up electrons within energy range of peak a and b can be explained by crystal field theory. The partial DOS spectrums for Fe$^T$(Fe$^O$) atoms looks similar in the case of YIG or BIG because yttrium orbitals and bismuth orbitals all energetically overlap with the conduction bands high than $\sim 4$ eV. Thus the electronic structure below 4 eV can be understood with crystal field theory and orbital mixing for iron and oxygen orbitals. For $\sim -4$ eV to $\sim 4$ eV the t$_{2g}$ orbitals all lie lower energy than e$_g$ orbitals lie. This backups the crystal field theory in the understanding of the electronic structure. However, crystal field theory can not explain the formulation of the band gap in both materials. The crystal field for iron atoms corresponds to the weak field limit for 6 electrons in the Fe$^T$ and Fe$^O$ d orbitals. This results in 4 spins/ion, which agree well with our calculated atomic magnetization. This also explains that the empty orbitals for Fe$^T$ site is purely spin down and that of Fe$^O$ site is purely spin up, as 6 spins corresponds to 4 unpaired spins and 2 paired spins. The difference in the energy of t$_{2g}$ and e$_g$ orbitals is larger in the case of BIG compare to that of YIG. It is mostly likely due to the structure difference, as yttrium d orbitals and bismuth p orbitals all energetically overlap with the conduction bands high than $\sim 4$ eV. One can see for the YIG scalar-relativistic DOS spectrum, there is a low-flat structure near CBM from $E = 1.8$ eV to $E = 2.5$ eV. This is caused by the first few bands near CBM, which lead to underestimated first main absorption edge as mentioned above.

For YIG DOS calculated in scalar-relativistic approximation, the numbers of the spin up valence bands and the spin down valence bands in primitive cell are quantized. This is why the difference of spin up valence bands number and the spin down valence bands number in primitive cell is quantized - in this case 20 spins per primitive cell. This corresponds to $5 \mu_B / f.u.$. When the SOC is turned on, the spin up states and spin down states mix with each other. Thus the calculated $m_s$ may not be exactly $5 \mu_B / f.u.$ But this effect is small since spin mixing mainly happen on degenerate states and that Y(Bi) Partial DOS mainly lie in the conduction band. Substituting yttrium with bismuth did not change much in trend for the partial DOS below Fermi energy. Since bismuth and yttrium orbitals does not mix much with valence band, the magnetic structure of YIG and BIG are thus the same.

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**FIG. 6.** Calculated optical conductivity of Y$_3$Fe$_5$O$_{12}$. (a) Real part and (b) imaginary part of the diagonal element; (c) imaginary part and (d) real part of the off-diagonal element. All the spectra have been convoluted with a Lorentzian of 0.3 eV to simulate the finite quasiparticle lifetime effects. Red lines are the optical conductivity derived from the experimental dielectric constant. [14]
The $\sigma^{1}_{xy}$ and $\sigma^{2}_{xy}$ for both YIG and BIG share no similar feature except that all the spectra have oscillatory behaviors and $\sigma^{2}_{xy}$ is $\sim 0$ below absorption edge. The $\sigma^{2}_{xy}$ spectrum from YIG has large positive peaks at $\sim 3.4$ eV and $\sim 5.6$ eV and large negative peak at $\sim 4.3$ eV. The $\sigma^{2}_{xy}$ spectrum from BIG has large positive peaks at $\sim 3.0$ eV and $\sim 4.6$ eV and $\sim 6.5$ eV and large negative peak at $\sim 2.3$ eV and $\sim 4.0$ eV and $\sim 6.0$ eV and $\sim 7.3$ eV. The $\sigma^{1}_{xy}$ spectrum from YIG has large positive peaks at $\sim 4.8$ eV and large negative peak at $\sim 3.9$ eV and $\sim 7.0$ eV. The $\sigma^{1}_{xy}$ spectrum from BIG has large positive peaks at $\sim 2.5$ eV and $\sim 4.3$ eV and large negative peak at $\sim 2.0$ eV and $\sim 3.6$ eV and $\sim 5.7$ eV and $\sim 7.0$ eV.

We have mentioned before that the theoretical calculated YIG band gap does not agree with experimental measured band gap. One can see that the calculated $\sigma^{1}_{xx}$ of YIG starts to increase at $\sim 2.4$ eV. This agrees with the experimental band gap of the system. The $\sigma^{1}_{xx}$ of YIG is near 0 for photons below $\sim 2.4$ eV even though the calculated band gap is $\sim 1.8$ eV. This is because of the highly dispersive bands near CBM does not contribute much to the joint density of states below $\sim 2.4$ eV. As for BIG, no such highly-dispersive bands appear at CBM, thus the calculated band gap agrees roughly with the experimental measured band gap.

In order to compare with the available experimental data, we plot the experimental optical conductivity spectra in Figs. 6 and 7. The theoretical predicted diagonal part of optical conductivity tensor for both YIG and BIG match perfectly with that of the experimental measurements within the measured energy range.

On the other hand, the calculated $\sigma^{1}_{xy}$ and $\sigma^{2}_{xy}$ for YIG are not agreeing so well with the experimental measured data. For $\sigma^{1}_{xy}$ spectrum, there is a peak at $\sim 4.8$ eV in the experimental measured data while no such peak is found in the theoretical calculated $\sigma^{1}_{xy}$ spectrum near $\sim 4.8$ eV. For $\sigma^{2}_{xy}$ spectrum, there is a peak at $\sim 4.5$ eV in the experimental measured data while no such peak is found in the theoretical calculated $\sigma^{1}_{xy}$ spectrum near $\sim 4.5$ eV. However, the trend of the calculated $\sigma^{1}_{xy}$ ($\sigma^{2}_{xy}$) is the same as that of the experimental measured data [14].

As for BIG, the calculated $\sigma^{1}_{xy}$ and $\sigma^{2}_{xy}$ agree well with the experimental measured data. The peaks and valleys in the measured $\sigma^{1}_{xy}$ and $\sigma^{2}_{xy}$ spectra are also found in our theory. The peak positions, peak heights and overall trend of experimental data match to that of our theory.

The optical selection rule for irreps at $\Gamma$ point is listed in the supplement material [S1]. The transitions between states at $\Gamma$ point can only be left(right) circular polarized or z-linear polarized, as restricted by symmetry of the solids. We expect $\Gamma$ point will give large contribution to the spectrum as band extremum often happen at $\Gamma$ point. Therefore we analyze the band transition accordingly to the selection rule and try to find out the main transition in both systems. However the symmetry allowed transitions are too many and cannot give firm conclusions. In order to find out the dominant transitions in Eq. (3), we calculate and compare the transi-

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**FIG. 7.** Calculated optical conductivity of Bi$_3$Fe$_5$O$_{12}$. (a) Real part and (b) imaginary part of the diagonal element; (c) imaginary part and (d) real part of the off-diagonal element. All the spectra have been convoluted with a Lorentzian of 0.3 eV to simulate the finite quasiparticle lifetime effects. Red lines are the optical conductivity derived from the experimental dielectric constant. [13]
tion matrix elements $Im(p_{ij}^x p_{ji}^y)$ for the transitions and listed out the transitions that is dominating and relevant (transitions with $|Im(p_{ij}^x p_{ji}^y)| > 0.12$ eV for BIG; transitions with $|Im(p_{ij}^x p_{ji}^y)| > 0.10$ eV for YIG). The main transition is presented in Fig. 8,9. They correspond to the peaks in the absorptive spectrum of $\sigma_{xy}$.

The band symmetries corresponding to the main transitions at $\Gamma$ point is listed at supplement material [S2]. In the case of YIG, N1-P1, N2-P2 and N3-P4 are paired transitions; while for BIG, all main transitions are not paired. We highlight that for BIG, N1-4 are all transitions near band gap. The left circular nature for the transitions near band gap is explained in [39] as a result of BIG being a single spin semiconductor and that bismuth orbitals mostly mix with unoccupied states. The reason that most main transitions are paired in the case of YIG is because spin-orbit coupling only slightly perturbed the band diagram. As one would expect without spin-orbit coupling there will be no MO effect without the inclusion of spin-orbit coupling, therefore YIG will not posses MO effect at the band gap can be estimated from the full-relativistic band diagram of BIG, which is roughly $0.17 \sim 0.32$ eV (supplement material[S3]). Eq. (9) can be verified in Fig. 10. as we plotted $\delta \omega$ at the band gap. The large negative Kerr rotation maximum occurs at the photon energy of $\sim 3.6$ eV(0.10°) for the bulk YIG, of $\sim 3.5$ eV(0.80°) for the bulk BIG. The large Kerr ellipticity maximum occurs at the photon energy of $\sim 4.1$ eV(0.16°) for the bulk YIG, of $\sim 1.9$ eV(0.54°) for the bulk BIG. On the other hand, the large negative Kerr rotation maximum occurs at the photon energy of $\sim 4.8$ eV(-0.12°) for the bulk YIG, of $\sim 2.4$ eV(-1.21°) for the bulk BIG. The large negative Kerr ellipticity maximum occurs at the photon energy of $\sim 3.3$ eV(-0.13°) for the bulk YIG, of $\sim 2.9$ eV(-1.16°) for the bulk BIG.

D. Magneto-optical Kerr and Faraday effect

Here we study the polar Kerr and Faraday effect for YIG and BIG. The complex Kerr and Faraday rotation angles as a function of photon energy for YIG and BIG are plotted in Figs. 8 and 9. The overall trend for the calculated MOKE spectra for YIG and BIG are quite different except for the oscillatory behavior. The large Kerr rotation maximum occurs at the photon energy of $\sim 3.6$ eV(0.10°) for the bulk YIG, of $\sim 3.5$ eV(0.80°) for the bulk BIG. The large Kerr ellipticity maximum occurs at the photon energy of $\sim 4.1$ eV(0.16°) for the bulk YIG, of $\sim 1.9$ eV(0.54°) for the bulk BIG. On the other hand, the large negative Kerr rotation maximum occurs at the photon energy of $\sim 4.8$ eV(-0.12°) for the bulk YIG, of $\sim 2.4$ eV(-1.21°) for the bulk BIG. The large negative Kerr ellipticity maximum occurs at the photon energy of $\sim 3.3$ eV(-0.13°) for the bulk YIG, of $\sim 2.9$ eV(-1.16°) for the bulk BIG.
We can compare our calculated Kerr rotation angles with other known MO material families such as 3d transition metal alloy and compound semiconductors. For magnetic metals, ferromagnetic 3d transition metals and their alloys are an important family. Among them, manganese-based pnictides are known to have strong MO properties. In particular, MnBi thin films were reported to have a large Kerr rotation angle of 2.3° (at photon energy ~ 1.84 eV) [36, 41] Pt alloys such as FePt, CoPt [42] and PtMnSb [43] possess large Kerr rotation angles. It was shown that the strong SOC of heavy Pt in these systems are important for the cause of strong MOKE. [42] Among semiconductor MO materials, diluted magnetic semiconductors Ga1-xMnxAs were reported to show large Kerr rotations angle as large as ~ 0.4 ° (of photon energy ~ 1.80 eV) [44] Therefore, the strong MOKE effect in YIG and BIG could have promising applications such as high density MO data-storage devices or MO nanosensors with high spatial resolution. YIG and BIG are also known to have excellent microwave properties (low magnetic damping) and a perfect spin current conductor. YIG and BIG may have applications in spintronics devices as interfaces between microwave, spin current and optics.

The calculated complex Faraday rotation angles for YIG and BIG film are presented in Fig. 9. The large Faraday rotation maximum occurs at the photon energy of ~ 3.9 eV (7.2°/µm) for the YIG, of ~ 3.7 eV (51.2°/µm) for the bulk BIG. The large Faraday ellipticity maximum occurs at the photon energy of ~ 4.4 eV (7.9°/µm) for the bulk YIG, of ~ 2.3 eV (54.1°/µm) for the bulk BIG. On the other hand, the large negative Faraday rotation maximum occurs at the photon energy of ~ 5.4 eV (-5.7°/µm) for the bulk YIG, of ~ 2.7 eV (-74.6°/µm) for the bulk BIG. The large negative Faraday ellipticity maximum occurs at the photon energy of ~ 6.6 eV (-5.6°/µm) for the bulk YIG, of ~ 3.2 eV (-70.2°/µm) for the bulk BIG. For comparison, we notice that MnBi films are known for large Faraday rotation angles of ~ 80°/µm (at 1.8 eV). [36, 41] Finally, we compare our predicted MOKE and MOFE spectra with experimental measurements within available photon energy range. All of predicted MOKE and MOFE are in good agreement with experimental measurements [14, 12, 14, 15]. Our theoretical predictions will have better agreement if all of the calculated spectra are blue-shifted by 0.3 eV. The measured MOKE and MOFE angle are larger than our theory but within acceptable range.

IV. CONCLUSION

To summarize, we have systematically studied the magnetism, electronic structure, optical and MO properties of cubic iron garnets Y3Fe5O12 and Bi3Fe5O12 by performing GGA+U calculations. We find that both Y3Fe5O12 and Bi3Fe5O12 exhibit strong MO effects. In particular, the Kerr rotation angle of Bi3Fe5O12 becomes as large as -1.2° at photon energy ~ 2.4 eV, and the Faraday rotation angle for BIG film reaches -74.6 °/µm at photon energy ~ 2.7 eV. The principal features in the optical and MO conductivity spectra are analyzed in terms of the calculated band structures and especially the interband optical transition matrix elements at the Γ point. For example, we find that the main peak of YIG and BIG
MO conductivity spectra are caused by different transitions. Thus the enhanced MO properties due to substitution of Y in YIG with Bi cannot be understood as merely enhancement of the SOC in the mixing of heavy Bi atomic orbital, while structure difference and orbital mixing of heavy element yttrium (bismuth) should also be taken account. We found that the strong MO effect near band gap of BIG is due to its nature of being a single-spin semiconductor and having bismuth orbital mixed mainly in conduction band. Our findings of strong MO effects in both iron garnets and also single-spin semiconductivity in Bi$_3$Fe$_5$O$_{12}$ suggest that the iron garnets are a playground of exploring the interplay of microwave, spin current, magnetism, and optics degrees of freedom and also have promising applications in high density semiconductor MO and low-power spintronics nanodevices, or as playground of coupling microwave, spin current, magnetism, and optics degree of freedom altogether.

FIG. 11. Calculated complex Kerr rotation angle. (a) Kerr rotation ($\theta_K$) and (b) Kerr ellipticity ($\varepsilon_K$) spectra of Y$_3$Fe$_5$O$_{12}$; (c) Kerr rotation ($\theta_K$) and (d) Kerr ellipticity ($\varepsilon_K$) spectra of Bi$_3$Fe$_5$O$_{12}$. Red circles in (a) and (b) denote the experimental values from Ref. [15].

FIG. 12. Calculated complex Faraday rotation. (a) Faraday rotation ($\theta_F$) and (b) Faraday ellipticity ($\varepsilon_F$) spectra of Y$_3$Fe$_5$O$_{12}$; (c) Kerr rotation ($\theta_K$) and (d) Kerr ellipticity ($\varepsilon_K$) spectra of Bi$_3$Fe$_5$O$_{12}$. Red dashed line in (a) denotes the measured values from Ref. [15]. Black circles in (a) and (b) are the experimental values from Ref. [45]. Red (green) circles in (c) and (d) are the experimental values from Ref. [45] (17).
[1] V. Cherepanov, I. Kolokolov, and V. Lvov, "The saga of YIG: spectra, thermodynamics, interaction and relaxation of magnons in a complex magnet," Phys. Rep. 229, 81 (1993).

[2] S. Mizukami, Y. Ando, and T. Miyazaki, "Effect of spin diffusion on Gilbert damping for a very thin permalloy layer in Cu/permalloy/Cu/Pt films," Phys. Rev. B 66, 104413 (2002).

[3] S. Chikazumi, *Physics of Ferromagnetism*, 2nd ed. (Oxford University Press, Oxford, 1997).

[4] Y. Kajiwara, K. Harri, S. Takahashi, J. Ohe, K. Uchida, M. Mizuguchi, H. Umezawa, H. Kawai, K. Ando, K. Takahashi, S. Maekawa, and E. Saitoh, "Transmission of electromagnetic signals by spin-wave interconversion in a magnetic insulator, Nature (London) 464, 262 (2010).

[5] T. Schneider, A. A. Serga, B. Leven, B. Hillebrands, R. L. Stamps, and M. P. Kostylev, "Realization of spin-wave logic gates," Appl. Phys. Lett. 92, 022505 (2008).

[6] Y. Sun, H. Chang, M. Kabatek, Y.-Y. Song, Z. Wang, M. Jantz, W. Schneider, M. Wu, E. Montoya, B. Kardasz, B. Heinrich, S. G. E. te Velthuis, H. Schultheiss, and A. Hoffmann, "Damping in Yttrium Iron Garnet Nanoscale Films Capped by Platinum," Phys. Rev. Lett. 111, 106601 (2013).

[7] P. M. Oppeneer, Chapter 1 Magneto-optical Kerr Spectra, pp. 229-422, in *Handbook of Magnetic Materials*, edited by K. H. J. Buschow. Elsevier, Amsterdam, (2001).

[8] V. Antonov, B. Harmon, and A. Yaresko. *Electronic structure and magneto-optical properties of solids*. Springer Science & Business Media, (2004).

[9] J. P. Castera, in *Magneto-optical Devices*, Vol. 9 of Encyclopedia of Applied Physics, edited by G. L. Trigg (Wiley-VCH, New York, 1996), p. 133.

[10] M. Mansuripur, *The Principles of Magneto-Optical Recording* (Cambridge Univ. Press, Cambridge, 1995).

[11] F. D. M. Haldane and S. Raghu, "Possible Realization of Spin-Induced Magneto-Optical Devices," Phys. Rev. B 87, 4867 (2000).

[12] T. Okawa, S. Suzuki, and K. Nakao, "First-principles study of spin-orbit interactions in bismuth iron garnet," J. Phys. Soc. Jpn. 74, 401 (2005).

[13] B. Hoffmann, Damping in Yttrium Iron Garnet Nanoscale Films Capped by Platinum, Phys. Rev. Lett. 111, 106601 (2013).

[14] H. Toraya and T. Okuda, "Crystal structure analysis of polycrystalline Bi$_2$Fe$_4$O$_{12}$ thin film by using asymmetric and symmetric diffraction techniques," J. Phys. Chem. Solids 56, 1317 (1995).

[15] J. P. Perdew, K. Burke, and M. Ernzerhof, "Generalized Gradient Approximation Made Simple," Phys. Rev. Lett. 77, 3865 (1996).

[16] S. L. Dudarev, G. A. Botton, S. Y. Savrasov, C. J. Humphreys, and A. P. Sutton, "Electron-energy-loss spectra and the structural stability of nickel oxide: An LSDA+ U study," Phys. Rev. B 57, 1505 (1998).

[17] H.-T. Jeng, G. Y. Guo and D. J. Huang, "Charge-orbital ordering and Verwey transition in magnetite," Phys. Rev. Lett. 93, 156403 (2004).

[18] G. Kresse and D. Joubert, "From ultrasoft pseudopotentials to the projector augmented-wave method," Phys. Rev. B 59, 1758 (1999).

[19] G. Kresse and J. Furthmüller, "Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set," Phys. Rev. B 54, 11169 (1996).

[20] G. Kresse and J. Furthmüller, "Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set," Comput. Mat. Sci. 6, 15 (1996).

[21] W. Feng, G.-Y. Guo, J. Zhou, Y. Yao, and Q. Niu, "Large magneto-optical Kerr effect in noncollinear antiferromagnets Mn$_3$X (X = Rh, Ir, Pt)" Phys. Rev. B 92, 144426 (2015).

[22] C. S. Wang and J. Callaway, "Band structure of nickel: Spin-orbit coupling, the Fermi surface, and the optical conductivity," Phys. Rev. B 9, 4897 (1974).

[23] P. M. Oppeneer, T. Maurer, J. Sticht, and J. Kbler, "Optical magnetooptical Properties of Single-Crystal Orthoferrites, Garnets, and Other Ferric Oxide Compounds," Phys. Rev. 186, 891 (1969).

[24] W. Chen, F. -Y. Lo, D. -R. Liu, K. Yang, and J. -S. Liaw, "Red shift of Faraday rotation in thin films of completely bismuth-substituted iron garnet Bi$_2$Fe$_5$O$_{12}$," Jpn. J. Appl. Phys., Part 1 38, 6687 (1999).

[25] E. Jesenska, T. Yoshida, K. Shinozaki, T. Ishibashi, L. Beran, M. Zahradnik, R. Antos, M. Kucera, and M. Veis, "Optical and magnetooptical properties of Bi substituted yttrium iron garnets prepared by metal organic decomosition, Opt. Mater. Express 6(6), 19861997 (2016).
[36] P. Ravindran, A. Delin, P. James, B. Johansson, J. Wills, R. Ahuja, and O. Eriksson, Magnetic, optical, and magneto-optical properties of MnX (X=As, Sb, or Bi) from full-potential calculations, Phys. Rev. B 59, 15680 (1999).

[37] D. Rodic, M. Mitric, R. Tellgren, H. Rundlof, and A. Kremenovic, True magnetic structure of the ferrimagnetic garnet Y₃Fe₅O₁₂ and magnetic moments of iron ions, J. Magn. Magn. Mater. 191, 137 (1999).

[38] N. Adachi, T. Okuda, V. P. Denysenkov, A. Jalali-Roudsar, and A. M. Grishin, Magnetic properties of single crystal film Bi₃Fe₅O₁₂ prepared onto Sm₃(Sc,Ga)₅O₁₂(1 1 1), J. Magn. Magn. Mater. 242-245, 775 (2002).

[39] M. Deb, E. Popova, A. Fouchet, and N. Keller, Full spin polarization of complex ferrimagnetic bismuth iron garnet probed by magneto-optical Faraday spectroscopy, Phys. Rev. B 87, 224408 (2013).

[40] N. Nagaosa, J. Sinova, S. Onoda, A. H. MacDonald, and N. P. Ong, Anomalous Hall effect, Rev. Mod. Phys. 82, 1539 (2010).

[41] G. Q. Di and S. Uchiyama, Optical and magneto-optical properties of MnBi film, Phys. Rev. B 53, 3327 (1996).

[42] G. Y. Guo and H. Ebert, On the origins of the enhanced magneto-optical Kerr effect in ultrathin Fe and Co multilayers, J. Magn. Magn. Mater. 156, 173 (1996).

[43] P. Van Engen, K. Buschow, R. Jongebreur, and M. Erman, PtMnSb, a material with very high magneto-optical Kerr effect, Appl. Phys. Lett. 42, 202–204 (1983).

[44] R. Lang, A. Winter, H. Pascher, H. Krenn, X. Liu, and J. K. Furdyna, Polar Kerr effect studies of Ga₁₋ₓMnₓAs epitaxial films, Phys. Rev. B 72, 024430 (2005).

[45] M. Deb, E. Popova, A. Fouchet, and N. Keller, Magneto-optical Faraday spectroscopy of completely bismuth-substituted Bi₃Fe₅O₁₂ garnet thin films, J. Phys. D 45, 455001 (2012).