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ABSTRACT

The anisotropic optical absorption edge of β-Ga_2O_3 follows a modified Beer–Lambert law having two effective absorption coefficients. The absorption coefficient of linearly polarized light reduces to the least absorbing direction beyond a critical penetration depth, which itself depends on polarization and wavelength. To understand this behavior, a Stokes vector analysis is performed to track the polarization state as a function of depth. The weakening of the absorption coefficient is associated with a gradual shift of linear polarization to the least absorbing crystallographic direction in the plane, which is along the a-exciton within the (010) plane or along the b-exciton in the (001) plane. We show that strong linear dichroism near the optical absorption edge causes this shift in β-Ga_2O_3, which arises from the anisotropy and spectral splitting of the physical absorbers, i.e., excitons. The linear polarization shift is accompanied by a variation in the ellipticity due to the birefringence of β-Ga_2O_3. Analysis of the phase relationship between the incoming electric field to that at a certain depth reveals the phase speed as an effective refractive index, which varies along different crystallographic directions. The critical penetration depth is shown to be correlated with the depth at which the ellipticity is maximal. Thus, the anisotropic Beer–Lambert law arises from the interplay of both the dichroic and birefringent properties of β-Ga_2O_3.

The Beer–Lambert (BL) law states that in an absorbing medium, the photon flux (Φ) decays exponentially with penetration depth (z), $\Phi = \Phi_0 e^{-\alpha z}$, where Φ_0 is the photon flux at z = 0 and the exponent α is the medium's absorption coefficient.[1] However, the BL law is only applicable for materials with plane wave electro-magnetic (EM) eigenmodes whose polarization states do not alter during propagation. For highly anisotropic materials, such as monoclinic β-Ga_2O_3, this is not the case, and the BL law must be modified to include a dependence on the polarization angle (θ), penetration depth (z), and energy (E_{ph}) of the photon, called the anisotropic BL law,[2]

$$\frac{\Phi}{\Phi_0} = \begin{cases} e^{-\alpha z}, & 0 < z \le z_c \\ e^{-\alpha z_c} e^{-\alpha^*(z-z_c)}, & z_c < z \end{cases}, \tag{1}$$

where the flux decay follows the isotropic BL law with an absorption coefficient α until a critical penetration depth z_c is reached. Both α and z_c depend on the incoming wave's polarization angle θ and photon energy E_{ph}, respectively. Beyond z_c, α weakens to the smallest value of absorption coefficient α^* that is possible in the plane containing the EM fields. This is observed both in the a-c (010) and a-b (001) planes of β-Ga_2O_3 for the energy ranges 4.65–5.2 and 4.9–5.8 eV, respectively. To understand the origin of this phenomenon, first described in Ref. 2, here, we study the evolution of the polarization of light propagating within β-Ga_2O_3. It will be shown that the polarization of light varies as it travels into the material due to dichroism and birefringence.

A monochromatic EM wave consists of a wave vector k, a phase, and a polarization state, which is generally described by four parameters: intensity (I), degree of polarization (p), and the shape parameters (ψ, χ) of the polarization ellipse. The Stokes vector is a set of values that describe the polarization state of an EM wave.[3,4] The relationship of the Stokes vector values to polarization state parameters is given as follows:[5,6] $S_0 = I$, $S_1 = Ip\cos 2\psi \cos 2\chi$, $S_2 = Ip \sin 2\psi \cos 2\chi$, and $S_3 = Ip \sin 2\chi$. Given the Stokes vector values, the polarization state parameters are $I = S_0$, $p = \frac{\sqrt{S_1^2+S_2^2+S_3^2}}{S_0}$, $2\psi = \tan^{-1}\frac{S_2}{S_1}$, and

$2\chi = \tan^{-1}\frac{S_3}{\sqrt{S_1^2+S_2^2}}$. The linear polarization direction is controlled by ψ, while χ determines the change of ellipticity (linear to elliptical to circular to elliptical to linear) of the polarization state of the EM wave. In a fixed Cartesian $(\hat{x},\hat{y})$ basis, $S_0 = |E_x|^2 + |E_y|^2$, $S_1 = |E_x|^2 - |E_y|^2$, $S_2 = 2\,Re(E_xE_y^*)$, and $S_3 = -2\,Im(E_xE_y^*)$, given E_x and E_y are the complex components of the electric field, i.e., $\vec{E} = \hat{x}E_x + \hat{y}E_y$.[5,6] The details of the EM calculations and geometry with respect to the crystallographic planes of $\beta - Ga_2O_3$ were provided, respectively, in the Appendix and Fig. 1 of Ref. 2. The polarization state at any depth can be determined by tracking the evolution of the Stokes vector. For light linearly polarized along the x direction, i.e., $|E_x| = 1$, $|E_y| = 0$, the initial value of $\psi = \theta$ and $\chi = 0°$.

Figure 1 shows the changes in ψ and χ for light incident on the a-b (001) plane as a function of depth for various E_{ph} and θ. The a-axis is along $\theta = 0°$, and the b-axis is along $\theta = 90°$, while the other two angles represent non-major axes directions. For the major axes, $\psi = \theta = 0°$ for the a-axis, $\psi = \theta = 90°$ for the b-axis, and $\chi = 0$ for all z. For all other directions, ψ shifts toward 90°, the b-axis, as it propagates. The b-axis in the a-b (001) plane is the dipole moment direction of the b-exciton (X_b),[7,8] and, thus, light in this energy range becomes aligned to the dipole moment of X_b after traversing a length of ∼3 μm in the material. This is true for all other linear polarizations that are not aligned with the major axes in the (001) plane. A phase shift, ellipticity, is introduced just within the surface $(z > 0)$, and χ moves away from 0° toward positive or negative values for non-major axes directions. After a range of $z \sim 3000$ nm, χ returns to 0°, which reveals that the ellipticity is not permanent, and beyond a certain depth ∼3 μm, the light will again be fully linearly polarized along the b-axis.

Figure 2 shows the changes in ψ and χ for light incident on the a-c (010) plane at various E_{ph} and θ. The a-axis is along $\theta = 0°$, the c-axis is along $\theta = 103.8°$, $\theta = 25.2°$ is along the dipole moment of the a-exciton (X_a), and $\theta = 115.1°$ is parallel with the c-exciton (X_c).[7,8] Unlike the a-b (001) plane, all directions including the major axes in a-c (010) plane experience variations in ψ and χ. For all incoming polarization states, ψ shifts to $20° - 25°$ at a depth of $z \sim 10\,\mu$m. For $4.9\,\text{eV} \le E_{ph} < 5.2\,\text{eV}$, $\psi \sim 25°$ but in the range $E_{ph} < 4.9\,\text{eV}$, ψ approaches ∼20°. Just like the a-b (001) plane, for all directions in the a-c (010) plane, the ellipticity variation is introduced just within the surface $(z > 0)$, and χ starts to move away from 0° for all directions. However, the ellipticity variation parameter never settles back to $\chi = 0°$, rather has some residual value even at 10 μm for all directions. Thus, initially linearly polarized light incident on β-Ga_2O_3 becomes elliptical when the electric field of the traveling wave is in the a-c (010) plane. Nevertheless, for all directions, χ remains very close to 0° (within ±1°) for $4.8\,\text{eV} \le E_{ph} \le 5\,\text{eV}$ when light has traveled deep enough, $z \ge \sim 10\,\mu$m. Mostly linear polarized light is recovered along the dipole moment of X_a in the a-c (010) plane at a depth of ∼10 μm for this energy range.

In the anisotropic BL law for β-Ga_2O_3, the value of α^* was given as $\alpha^* = \alpha(\theta = 90°)$ in the a-b (001) plane and the $\alpha^* = \alpha(\theta \cong 25°)$ in the a-c (010) plane. It is now established that the incoming wave's linear polarization state is shifted toward a specific direction in each plane of $\beta - Ga_2O_3$. These directions are oriented along $\theta = 90°$ in the a-b (001) and $\theta \cong 20°$–$25°$ in the a-c (010) planes. The values of α^* match α along these directions in the anisotropic BL law. Moreover, these directions are parallel with the exciton dipole moments in β-Ga_2O_3, which are oriented along X_b at 90° from the a-axis in the a-b (001) plane and along X_a at a range of ∼17°–25.2° from the a-axis in the a-c (010) plane. The third exciton X_c is oriented at an angle in the range ∼110°–115.1° from the a-axis in the a-c (010) plane.[7,8] These excitonic transitions are the three lowest near bandgap critical point

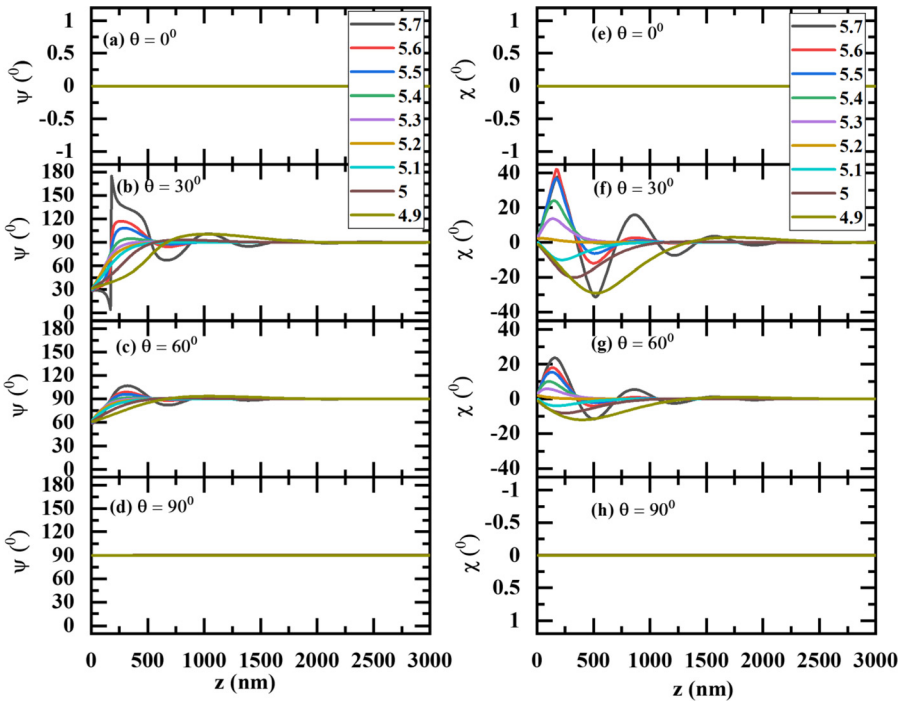

FIG. 1. (a) and (d) Linear polarization shape parameter ψ and (e)–(h) ellipticity shape parameter χ for light polarized within the a-b (001) plane with initial polarization direction θ, incident on β-Ga_2O_3, and propagating to a depth z for different photon energies (shown in the legends in units of eV). The polarization state for light along $\theta = 0°$ a- and $\theta = 90°$ b-axes does not vary with z; however, there is a gradual shift of ψ toward the b-axis for other polarization states as well as ellipticity introduced near the surface, which decays with depth.

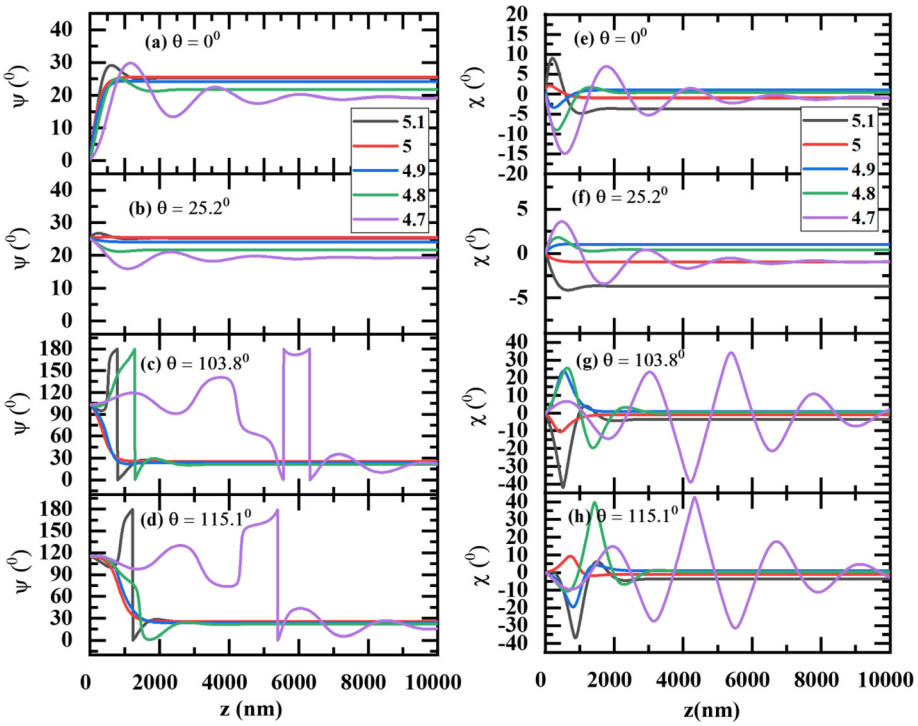

FIG. 2. (a)–(d) Linear polarization shape parameter ψ and (e)–(h) ellipticity shape parameter χ for light polarized within the a-c (010) plane with initial polarization direction θ, incident on β-Ga_2O_3, and propagating to a depth z for different photon energies (shown in the legends in units of eV). All polarization directions show a change in the polarization direction and develop ellipticity. The final linear polarization state shifts toward the direction if the dipole moment of the X_a exciton at $\psi \sim 20^\circ - 25^\circ$. The ellipticity persists to larger depths in the a-c (010) plane than in the a-b (001) plane (Fig. 1) before settling near 0° for most of the energies in the range of interest.

transitions, including excitonic contributions[7,8] that take place at energies $X_c \sim 4.9$ eV, $X_a \sim 5.2$ eV, and $X_b \sim 5.5$ eV. Since these transitions dominate the near bandgap photon absorption process[9,10] in β-Ga_2O_3, it is expected from the transition energies that in the a-b (001) plane, the least absorbing direction will be along X_b $(\theta = 90^\circ)$ in the energy range $4.9 - 5.8$ eV, and in the a-c (001) plane, the least absorbing direction will be along X_a $(\theta \cong 20^\circ - 25^\circ)$ in the energy range $4.65 - 5.2$ eV. Thus, the physical basis of weakening of the absorption coefficient to the smallest value possible in plane is established to be a result of the anisotropy and energy shift of the physical absorbers, i.e., excitons.

The gradual shift of ψ happens due to dichroism[2] of β-Ga_2O_3, which is the change in absorption of light with polarization direction.[11] Linear Dichroism (L_D) is defined as the difference in absorbance of light that is linearly polarized along orthogonal directions within a plane. Light aligned along any other polarization direction incident on the plane can be described by two components along these two orthogonal directions. Since one direction absorbs more than the other direction, the light component aligned along this more-absorbing direction will lose its amplitude faster than the other component. As a result, the sum of the components will gradually shift toward the less absorbing linear polarization direction.

A good illustration of this behavior is for linearly polarized light incident on the a-b (001) plane in β-Ga_2O_3. The two orthogonal directions are a- and b-axes, and the L_D in a-b (001) plane is shown in Fig. 5 of Ref. 2. Since non-major axes polarized light's a-axis component is absorbed more,[2] it will lose its amplitude faster than the component aligned along the b-axis, thus effectively shifting the non-major axes polarized light to the b-axis after propagating some distance. Similarly, in the a-c (010) plane, since the light is absorbed more along the X_c direction,[2] the non-dipole-moment-oriented polarized light will eventually shift to the X_a direction, which is nearly orthogonal to the X_c direction.[2] The L_D in the a-b (001) plane has higher magnitude at any given depth compared to the a-c (010) plane. So, the change of ψ to its final polarization state is completed earlier in the a-b (001) plane at a depth of ~3 μm compared to the a-c (010) plane at a depth of ~10 μm.

The shift of ψ is accompanied by a strong variation of χ. The depth required for the ellipticity to be removed and to recover a completely linear polarization state is larger for the a-c (010) plane compared to the a-b (001) plane. Even then, for the a-c (010) plane, the ellipticity is not completely removed rather has some residual value. The origin of this ellipticity parameter variation lies in the birefringence property[12,13] of β-Ga_2O_3. Birefringence is defined as the change in refractive index with change in polarization and propagation directions.[14] Refractive index, n, of a material is directly related to the velocity of light in the material. The refractive index is the ratio of light's velocity in vacuum, c, to that in the material, v, i.e., $n = \frac{c}{v} = \frac{ck}{\omega}$, where ω is the angular frequency and k is the EM wave vector. Linear birefringence (L_B) is the difference in refractive index of linearly polarized light along orthogonal directions in a plane. Then, the L_B between two orthogonal directions 1 and 2 will be defined as $\Delta n = n_2 - n_1 = \frac{c(k_2 - k_1)}{\omega}$. This means L_B produces a relative difference in the wave vector of the orthogonal components of the wave while passing through the plane. As a result, in one direction, light will move faster than the other, and as a result, there will be a retardation in phase angle between the orthogonal components aligned along these directions. Thus, a strong variation of the ellipticity parameter will be introduced.

To calculate the L_B, we need to find the refractive index along different directions in β-Ga_2O_3. The refractive index of a thin layer can be obtained by examining the phase relationship between the

incoming and outgoing electric field for a given polarization, utilizing the fact that the amount of phase retardation is related to the refractive index in the material. If the electric field for a linearly polarized light is given as $|E| = E_0 e^{-i(\omega t - kz)}$, then the phase of the field is $\varphi = \omega t - kz$. Just under the surface, any change in the electric field will be instantaneous, i.e., $t \sim 0$. If the wave propagates only a small distance, $\Delta z \sim 0$ within this $t \sim 0$ time, then the change in phase will be $\Delta\varphi = \varphi_{out} - \varphi_{in} = k(\Delta z - 0) = k\Delta z$. From this equation, the propagation vector can be obtained as $k = \frac{\Delta\varphi}{\Delta z}$. We know the velocity of light in a material is given as $v = \frac{\omega}{k} = \frac{\frac{E_{ph}}{\hbar}}{\frac{\Delta\varphi}{\Delta z}} = \frac{E_{ph}\Delta z}{\hbar\Delta\varphi}$. An effective refractive index can be obtained as $n^* = \frac{c}{v} = \frac{c\hbar\Delta\varphi}{E_{ph}\Delta z}$, where c is the velocity of light in vacuum and $\hbar$ is Planck's constant. Thus, considering propagation perpendicular to the surface only, the phase speed can be obtained as an effective refractive index when the amount of phase retardation of the electric field after propagating a small distance into the material is known.

Figure 3 shows the effective refractive index (n^*) along different directions in both planes, i.e., for the a-b (001) plane in Figs. 3(a)–3(c) and for the a-c plane (010) in Figs. 3(d)–3(f) at different small distances $\Delta z = 10$, 20, and 30 nm, respectively. Just below the bandgap[7,15] $E_g \sim 5.04$ eV, n^* is higher for the a-axis compared to the b-axis, while above the bandgap, n^* is higher for the b-axis compared to the a-axis. Thus, the fast axis in the a-b (001) plane is energy dependent; below the bandgap, it is the b-axis, while above the bandgap, it is the a-axis. Similarly, in the a-c (010) plane, below the bandgap, the a-axis is faster than the c-axis, while above the bandgap, the c-axis is faster than the a-axis. Thus, just below the bandgap, the order of the effective refractive index is $n_c^* > n_a^* > n_b^*$, but above the bandgap, it is $n_b^* > n_a^* > n_c^*$. In the Near Infrared (NIR) range, far below the bandgap, the a-axis has the smallest refractive index, i.e., the highest light velocity in both planes, which is consistent with previous studies.[12,16,17]

The near surface L_B is much larger in the a-b (001) plane than in the a-c (010) plane at any given distance $\Delta z \leq 30$ nm, as shown in Fig. 4, where the variation in L_B for the a-b (001) plane is defined as

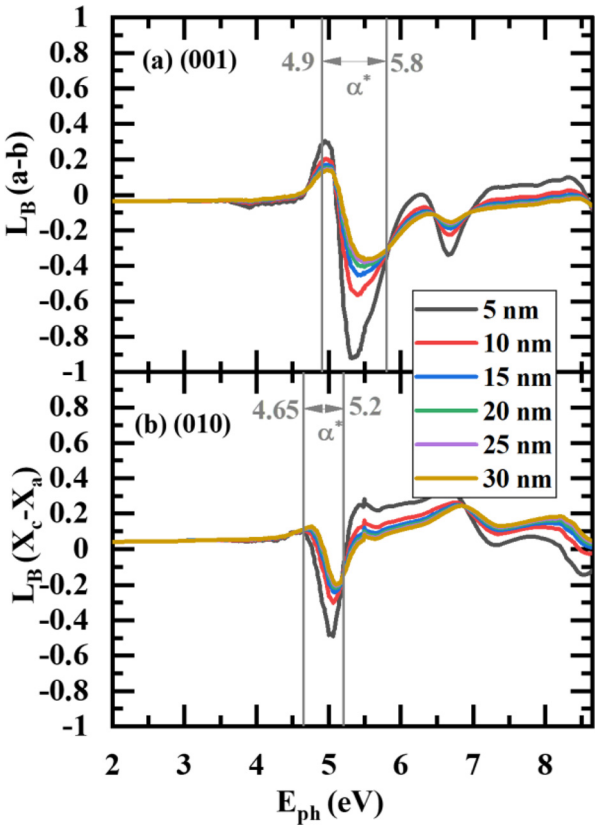

FIG. 4. (a) and (b) Photon energy E_{ph} dependence of the linear birefringence L_B for linearly polarized light incident on β-Ga_2O_3 within the (001) and the (010) planes, respectively, propagating a total depth of Δz (shown in the legend). L_B decays to 0 with increasing depth. The decay rate is larger for light polarized within the (001) plane compared with the (010) plane.

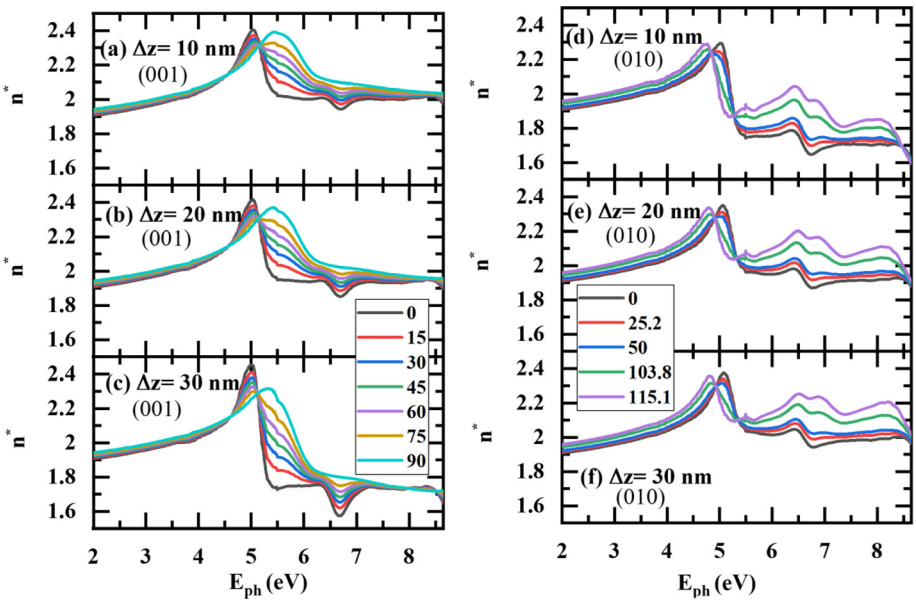

FIG. 3. Photon energy E_{ph} dependence of the effective refractive index (n^*) for linearly polarized light incident on β-Ga_2O_3 at an angle θ (shown in the legends in units of degrees) within the (001) plane in parts (a)–(c) or within the (010) plane in parts (d)–(f), after propagating a total depth of Δz. The n^* spectra are peaked near the bandgap for both planes. In the NIR range, the a-axis is always the fast axis regardless of the plane studied. In the vicinity of the excitonic transition energies, the n^* peaks broaden for the a-b (001) plane with increasing depth, while it narrows for the a-c (010) plane.

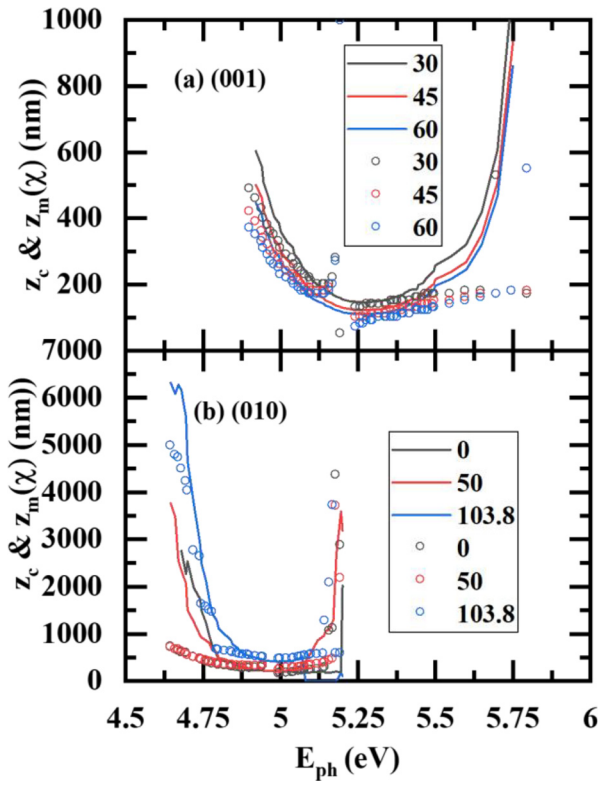

FIG. 5. (a) and (b) Photon energy E_{ph} dependence of the maximum ellipticity depth $z_m(\chi)$ (open symbols) compared with critical penetration depth z_c (lines) for linearly polarized light incident on β-Ga_2O_3 at an angle θ (shown in the legends in units of degrees) within the (001) plane and the (010) plane, respectively. $z_m(\chi)$ values are determined from the Stokes vector analysis results shown in Figs. 1 and 2, whereas z_c values are based on electromagnetic modeling of the Poynting vector decay, i.e., fits to the anisotropic BL law described in Ref. 2.

$\Delta n^* = n_a^* - n_b^*$ and as $\Delta n^* = n_{X_c}^* - n_{X_a}^*$ for the a-c (010) plane. In the energy range of interest, L_B is seen to be moving toward zero with increasing Δz for both planes. The maximum L_B appears closest to the surface and reduces as light travels deeper. So, at larger depths, β-Ga_2O_3 will act like an ideal linear polarizer with L_D effects only. This explains why the elliptical shape of the polarization state is introduced just below the surface but vanishes at a few micrometers depth. Since, near the surface, L_B is much higher, the initial change in χ is much larger in the a-b (001) plane compared to the a-c (010) plane. The rate of change in L_B is much higher in the a-b (001) plane compared to the a-c (010) plane. This is the reason for recovering zero ellipticity at a much smaller depth in the a-b (001) plane compared to that in the a-c (010) plane.

The ellipticity parameter variation maximizes at different depths for different incoming polarization states and photon energies. The critical penetration depth z_c was previously found to be polarization angle and energy dependent.[2] Figure 5(a) shows the depth, $z_m(\chi)$, at which χ is maximum, plotted as a function of energy for different polarization directions (dots) in the a-b (001) plane and compared with the critical penetration depth z_c in the same energy range and same directions (line). Figure 5(b) shows the same results for the a-c (010) plane. The critical penetration depth matches with the depth of maximal χ for both planes for the energy range $4.9\,\text{eV} \leq E_{ph} \leq 5.2\,\text{eV}$. For the a-b (001) plane, the results match even in the range $5.25\,\text{eV} \leq E_{ph} \leq 5.5\,\text{eV}$. Thus, for the energy range of the excitonic transitions, the depth at which χ is greatest, $z_m(\chi)$, corresponds with the critical penetration depth z_c. The depth of maximal χ corresponds with half the depth between the initial and final linear polarization state, thus, beyond this depth, the least absorbing direction's absorption coefficient α^* takes over the absorption process described by the anisotropic BL law.

The anisotropic BL law arises from the dichroism and birefringence properties of β-Ga_2O_3. While the dichroism property induces a change in the linear polarization direction, the birefringence property induces ellipticity. The linear polarization shift dictates the final linear polarization state, which, in turn, dictates the value of the weakened absorption coefficient in the anisotropic BL law. The critical penetration depth reflects the balancing point, beyond which the weaker absorption coefficient takes over and coincides with the maximal ellipticity. We also obtain the energy and polarization dependence of the effective refractive index, which provides the slow and fast axes over the wavelength range in β-Ga_2O_3 optoelectronic applications. Finally, the physical basis of the anisotropic absorption behavior in β-Ga_2O_3 is established to be originating from the anisotropy of the excitonic transitions. Such phenomena might have been overlooked and need thorough investigation in other materials with anisotropic excitons such as transition metal dichalcogenides and the entire class of low symmetry semiconductors.

See the supplementary material for plots of the three-dimensional (3D) Poincaré sphere representation, which is used to plot the evolution of the Stokes vector during propagation of light through Beta Gallium Oxide. The 3D plots are animated and provided for different atomic planes of incidence and initial polarization angles with respect to the lattice vectors.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Md Mohsinur Rahman Adnan: Formal analysis (lead); Investigation (lead); Methodology (lead); Writing – original draft (lead). **Mathias Schubert:** Formal analysis (supporting); Writing – review & editing (supporting). **Roberto C. Myers:** Conceptualization (equal); Formal analysis (equal); Investigation (lead); Methodology (equal); Supervision (lead); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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