



# Communication Non-Drude-Type Response of Photocarriers in Fe-Doped β-Ga<sub>2</sub>O<sub>3</sub> Crystal

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Abstract: Beta gallium oxide,  $\beta$ -Ga<sub>2</sub>O<sub>3</sub>, is one of the promising ultrawide bandgap semiconductors with a monoclinic (C2/m)  $\beta$ -phase structure showing strong anisotropic properties. To improve the performance of these devices, more optical characterization is required. Here, the anisotropic carrier dynamics in optically excited (010) oriented Fe-doped  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> was studied by terahertz time-domain spectroscopy. An 800 nm continuous-wave light source was employed to excite carriers from Fe acceptors to the conduction band and to probe an anisotropic interaction with the THz field. The complex THz conductivities fitted with the Drude and Drude–Smith models revealed that the optically excited carriers behave as nearly free carriers along the a-axis, whereas those along the c-axis show a non-Drude type response. The estimated mobility for the c-axis agrees with the reported values, whereas the results suggest much higher mobility along the a-axis.

Keywords: terahertz time-domain spectroscopy; Fe-doped beta gallium oxide; Drude and Drude–Smith models



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## 1. Introduction

β-Ga<sub>2</sub>O<sub>3</sub> attracts many scientists as an ultra-wide-bandgap semiconductor for applications such as gas sensors, power electronic devices, deep-ultraviolet (UV) photodetectors, and radiation detectors. The challenge lies in producing good-quality  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> and understanding its fundamental physical properties [1-6]. The resistivity of  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> varies between  $10^{-2}$  and  $10^{12} \Omega$  cm at room temperature (RT), and studies have revealed that impurities, doping, and band structures are critical for controlling its conduction [7–12]. Furthermore, owing to its inherently anisotropic crystalline structure [13–15],  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> demonstrates various anisotropic properties of the optical response, electrical conduction, bandgap, mobility, terahertz (THz) emission, and other parameters [16–24]; among these, anisotropic carrier transport is of great importance. Fe-doped  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> typically serves as a highly resistive semi-insulating substrate for device preparation [2,4] and can be a good platform for studying carrier dynamics with optical illumination. The  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> crystalline structure is drawn schematically in Figure 1a together with one of the possible locations of impurity (supplier of electrons) and Fe ion. There still remains intense discussion about impurity locations, and their energy levels such for unintentionally doped impurities, vacancies, self-trapped holes, and so on. Figure 1b gives the band diagram with the important impurities of this work. The materials include impurities such as Si, Sn, and Ge forming the shallow impurity levels, and the Fe ion in  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> works as an acceptor whose energy level is approximately 0.85 eV less than the conduction band minimum and which captures the electrons from donor-type impurities [9,25–27]. Illumination with an infrared laser can excite the photocarriers from those acceptors and realize a low carrier density in the semi-insulating  $\beta$ -Ga<sub>2</sub>O<sub>3</sub>.



**Figure 1.** (a) The unit cell of monoclinic  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> together with the possible locations of the impurity and Fe ion. The impurities work as shallow donors and provide free electrons nearby, and the Fe works as an acceptor and captures the thermally excited electron. (b) Energy band diagram for  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> with the shallow impurities and Fe acceptors.

Recently, THz time-domain spectroscopy (TDS) has become a powerful tool to discuss the carrier dynamics at THz frequencies and also estimate various DC parameters such as mobility  $\mu$ , carrier density N, carrier scattering time  $\tau$ , and DC conductivity  $\sigma_0$  in a non-contact manner. In comparison with conventional methods to characterize the semiconductor wafers such as a microwave photoconductivity decay (µ-PCD) [28], THz-TDS easily provides information on anisotropic properties and also non-Drude type carrier behaviour. THz-TDS is the tool to determine the refractive index of materials at THz frequencies [29,30]. In the present study, we examined the photocarrier behaviour in Fedoped (010)  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> using THz-TDS under CW light illumination. The refractive index is converted into dielectric constants and/or complex conductivities at the THz frequency range. One can easily estimate the various physical parameters of semiconductors, and THz-TDS is expected to provide useful information for the wide bandgap semiconductors to improve the quality and explore the carrier dynamics [31]. As for  $\beta$ -Ga<sub>2</sub>O<sub>3</sub>, recently, Blumenschein et al. reported the temperature dependence of THz properties and observed complicated carrier dynamics [32] and recently, Gopalan et al. studied the birefringence of  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> single crystals [33]. Thus, it is important to discuss the anisotropic THz properties of  $\beta$ -Ga<sub>2</sub>O<sub>3</sub>. However, THz-TDS has not been utilized yet for the study of photocarrier dynamics and is expected to provide new information on the anisotropic photocarrier dynamics THz properties of  $\beta$ -Ga<sub>2</sub>O<sub>3</sub>.

#### 2. Materials and Methods

The THz-TDS experiments in this study were carried out with a compact commercial THz-TDS (TR-1000, Otsuka Electronics Company Ltd., Hirakata, Japan), which employs an Er-doped femtosecond pulsed laser (FemtoliteCS-20, IMRA, Aisin, Kariya, Japan) at a repetition rate of 50 MHz and laser wavelength of 780 nm. The pulse width and average power of the laser at 780 nm were 65 fs and 25 mW, respectively. A laser diode with a wavelength of 800 nm was used to generate photocarriers in the Fe-doped  $\beta$ -Ga<sub>2</sub>O<sub>3</sub>. The THz radiation and CW laser light were focused onto the sample with a spot diameter of approximately 5 mm. We inject about 10<sup>15</sup> photons/cm<sup>3</sup>, which would not heat the samples. The experiments were performed at RT with a relative humidity of approximately 30–35%. Details of the experimental system and the illumination method have been previously

reported [34,35]. The sample used in this study was Fe-doped (010)  $\beta$ -Ga<sub>2</sub>O<sub>3</sub>, procured from Tamura Corporation, Japan and grown by the edge-defined film-fed method, with a thickness *d* of approximately 0.5 mm. To minimize data-related errors, the actual thickness of the sample was determined (492 µm) by minimizing the etalon effect using the first and second transmission pulses. (010)  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> exhibits birefringence along the a-axis and c-axis. Hence, we evaluated the effects of illumination on the complex conductivity with THz electric fields along the a-axis (E<sub>THz</sub>//a-axis) and c-axis (E<sub>THz</sub>//c-axis).

#### 3. Results and Discussion

Figure 2a shows the THz transmission waveforms through the Fe-doped (010)  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> with the THz fields  $E_{THz}$  along the a-axis and c-axis, which are clearly separated to estimate the birefringence. One can estimate the anisotropic refractive index which is given approximately by the following equations for optically thick samples.

$$n(\omega) = \frac{\varphi(\omega)c}{\omega d} + 1, \ \kappa(\omega) = -\frac{c}{\omega d} \ln \frac{\rho(\omega)(n+1)^2}{4n}$$
(1)

where  $\varphi(\omega)$  and  $\rho(\omega)$  are the amplitude and phase calculated from the Fourier transform of the data. Typically, for the thick samples, we use the first pulse only by removing the multiple reflections of the pulses. However, in the present work, we study the low conductivity with a small number of photo carriers, and then we use the first two pulses to minimize the estimation error. Figure 2b shows the anisotropic refractive index and dielectric constants of the unilluminated semi-insulating  $\beta$ -Ga<sub>2</sub>O<sub>3</sub>, which agrees with the previously reported values and we see no large etalon effects.

Laser illumination with a photon energy of 1.55 eV is expected to excite the photocarriers from the Fe acceptors into the conduction band and induce the absorption of the THz waves. Figure 3a shows the transmission waveforms at various powers. The second pulses were the waves that round-travelled once in the samples. Clear increases in the absorption were observed, indicating that the photocarriers were excited by illumination. The photon energy (1.55 eV) was much lower than the bandgap of  $\beta$ -Ga<sub>2</sub>O<sub>3</sub>. Although there has been a lack of research on the photoexcitation effect below the bandgap, optical absorption experiments show a finite value for the absorbance [17,27], suggesting that photocarrier excitation from the defect centres is attributed to absorption. According to studies on defects, we expect that the majority of the donors and acceptors in Fe-doped  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> are unintentionally doped impurities, vacancies of Ga and O, self-trap holes, and ionized Fe. With higher illumination power, more trapped electrons in the Fe sites are excited to the conduction bands, which then absorb the THz waves.





Figure 2. Cont.



(b)

**Figure 2.** (a) Waveforms of the THz pulses transmitted through the excited Fe-doped (010)  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> along E<sub>THz</sub>//a-axis and E<sub>THz</sub>//c-axis at 808 nm. (b) calculated refractive index and complex dielectric constants.



**Figure 3.** (a) Waveforms of the THz pulses transmitted through the excited Fe-doped (010)  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> along E<sub>THz</sub>//a-axis and E<sub>THz</sub>//c-axis at 800 nm with various illumination powers. (b) Calculated complex conductivity. The solid lines are obtained by using the first peak pulse only, and the dashed ones using the first and second pulses.

The conventional analytical method of THz-TDS [29] was used to estimate the complex conductivities,  $\tilde{\sigma}$ , using the complex refractive index,  $\tilde{n}$ , with the following equation:

$$\widetilde{n}^2 = \widetilde{\varepsilon} = \varepsilon_{\infty} - i \frac{\widetilde{\sigma}}{\omega \varepsilon_0}$$
<sup>(2)</sup>

where  $\tilde{\epsilon}$  is the complex dielectric constant,  $\epsilon_0$  is the permittivity of free space, and  $\epsilon_{\infty}$  is the dielectric constant at high frequencies.

Figure 3b shows the frequency dependence of the real and imaginary parts of the conductivity of the Fe-doped  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> (E<sub>THz</sub>//a-axis) with illumination at 808 nm with varying powers. The results indicate that the real part of the conductivity decreased with increasing frequency, and the imaginary part increased with increasing frequency in the THz range. and the real part of the conductivity increases with increasing illumination power, i.e., photocarrier density, whereas the small change was observed in the imaginary part. The solid lines represent the values estimated using the first pulse only, whereas the dashed ones represent the first two pulses. The latter ones include a large etalon effect, which mainly comes from misalignment of the THz wave paths and imperfection of the substrate surface parallelism. Note that one cannot avoid this kind of etalon effect for the poor conductive materials because THz-TDS has an intrinsic poor signal-to-noise ratio (~a few hundred) with a large beam diameter (~3 mm) [36]. Nevertheless, it is important to discuss the physics with the data considering the multiple reflections; as can be seen, there exist some discrepancies between the solid and dashed lines. Hence, the results will be discussed with double pulses taken into account.

The excited carriers are expected to behave as free carriers in the conduction band, and thus we calculated the fits to the data with the Drude model. In the Drude model, complex conductivity is defined as

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$$\tilde{\sigma} = \sigma_1 - i\sigma_2 = \frac{\sigma_0}{1 + \omega^2 \tau^2} - i\frac{\sigma_0 \omega \tau}{1 + \omega^2 \tau^2} \tag{3}$$

where  $\sigma_0$  is the DC conductivity,  $\tau$  is the effective scattering time, N is the carrier density, and  $m^*$  is the effective mass. The solid lines in Figure 4a are the calculated fits by a Drude model. The dashed lines are the fits by the Drude–Smith model. The Drude–Smith model and parameters will be discussed later. The overall trends of the frequency dependence were understood using a conventional Drude model.

Alternately, we found that the photoconductivity for the  $E_{THz}//c$ -axis deviated from the Drude model, as shown in Figure 4b. Small peaks existed at approximately 1 THz in the real part, whereas the imaginary part changed from positive to negative values. This type of behaviour is typically explained by the Drude–Smith model [37–39], whereby the carriers were localized in a small area or suffered from strong backscattering. The c-axis of (010)  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> is known to have a slightly smaller optical bandgap, whereas the a-axis has a larger bandgap. In this study, we excited a small number of electrons from the Fe acceptors, which are supposed to relax at the minimum energy level—this can be attributed to localization.

At higher frequencies, the deviation of the calculated lines from the data becomes large. This originates from the lowest phonon scattering modes, which exist around 6 THz, and have strong anisotropic properties [40]. The discussions are ongoing and should be studied by broadband THz-TDS with an air-plasma source [41]. We are currently working on that study.



**Figure 4.** (a) Real and imaginary parts of the conductivity of the Fe-doped (010)  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> (E<sub>THz</sub>//a-axis) at 800 nm with respect to various illumination powers; the solid lines are the fits with the Drude model, and the dashed ones are with the Drude–Smith model. (b) Real and imaginary parts of the conductivity of the Fe-doped (010)  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> (E<sub>THz</sub>//c-axis) at 800 nm with respect to various illumination powers from the front surface; the solid lines are the fits with the Drude–Smith model.

Various parameters can be estimated using the complex conductivity by either the Drude model or the Drude–Smith model. The carrier density  $N = \frac{m^*}{e^2 \tau} \sigma_0$  and the mobility  $\mu = \frac{e\tau}{m^*}$  are calculated by using the fitting parameters of  $\sigma_0$  and  $\tau$ , with the value of the effective mass  $m^* = 0.28 \text{ m}_0$  being the value of the electron effective mass that is most often obtained in both computation and experimentation at RT for  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> [14,42]. In the Drude–Smith model, the equations are modified as follows:

$$\tau_1 = \frac{ne^2\tau}{m^*(1+\omega^2\tau^2)} \left[ 1 + \frac{c(1-\omega^2\tau^2)}{(1+\omega^2\tau^2)} \right]$$
(4)

$$\sigma_2 = \frac{ne^2\tau}{m^*(1+\omega^2\tau^2)} \left[ 1 + \frac{2c}{(1+\omega^2\tau^2)} \right] \omega\tau$$
(5)

$$N = \frac{m^*}{e^2 \tau} \sigma_0 (1+c)^{-1} \tag{6}$$

$$\mu = \frac{e\tau}{m^*}(1+c) \tag{7}$$

where *c* is a measure of the persistence of velocity and can vary between 0 and -1. A negative value of *c* implies a predominance of backscattering; c = 0 corresponds to the Drude conductivity, and c = -1 corresponds to complete carrier backscattering or localization [43].

The estimated parameters are shown in Figure 5. Because a laser power of zero contains a large error due to its semi-insulating nature, it is excluded from the discussion. With increasing laser power, the carrier density and DC conductivity increased, as expected. The carrier density was approximately 50% higher for the  $E_{THz}//c$ -axis compared to the  $E_{THz}//a$ -axis. This is attributed to the smaller bandgap along the c-axis. Additionally, no significant dependence on mobility or scattering time was observed. However, a large

difference in the mobility along the a-axis and c-axis was found, whereas the scattering times were estimated to be approximately 100 fs. The values of the mobility along the c-axis are close to the observed values [8] and the theoretical maximum values [18]. It is difficult to provide a comprehensive explanation of the high mobility along the a-axis, but phenomenologically speaking, although the charges excite at the minimum conduction edge along the a-axis, the top of the saddle may not have strongly scattered from the self-trap holes and/or vacancy impurities. However, the anisotropy of the mobility was expected to be small [18]; our results may have been limited to the interaction of carriers with high-frequency electric fields.



**Figure 5.** Laser power dependence of excess carrier density, DC conductivity, mobility, and carrier scattering time. These parameters are estimated by fits with the Drude model ( $E_{THz}$ //a-axis) and Drude–Smith model ( $E_{THz}$ //c-axis).

It is also difficult to explain why the excited carriers interacted with the THz fields along the a-axis and c-axis as free carriers and localized/strongly backscattered carriers, respectively. Because we excited the carriers far above the minimum conduction band energy with the CW laser, we expected the carriers to be thermalized and to exist near the minimum band edge. The difference between the smaller and larger optical bandgap is approximately 30 meV, and thus, excited carriers may exist in both bands. Therefore, the band structure cannot explain the large difference in the anisotropic conduction model along the a-and c-axes. We need to consider the real-space transport of the high-speed carriers. One of the possible scenarios is that the electrons excited from the Fe impurities form the electron clouds at the specific location around the shallow donors, as depicted in Figure 1a, and/or floating charge states [44]. Those positions might be responsible for the strong anisotropic electron conduction; the electrons at the other positions may have more isotropic conduction properties. Regardless of the reason, it may be possible to realize faster devices by using electron travel in the a-axis direction, together with the reduction in shallow impurities.

### 4. Conclusions

In summary, we conducted a THz-TDS study on low-density carrier dynamics in optically excited Fe-doped  $\beta$ -Ga<sub>2</sub>O<sub>3</sub>. The 808 nm CW light source was employed to excite the carriers from the Fe acceptors into the conduction band and to probe the anisotropic interaction with the THz field. The complex THz conductivities fitted with the Drude and Drude–Smith models revealed that the optically excited carriers behave as free carriers along the a-axis, whereas those along the c-axis THz field show a non-Drude-type response. The results also suggest that the mobility along the a-axis is much higher than that along the c-axis, and the carriers suffer strong scattering and/or localization in the c-axis direction. We hope this study will inspire additional research on anisotropic high-speed carrier dynamics, which may contribute to the realisation of anisotropic power devices and high-frequency devices.

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